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Introduction

The ability to identify and accurately quantify uranium and plutonium is crucial to our national security. The protection, control, and accounting of these special nuclear materials play a central role in weapons dismantlement, storage, facility inspections, waste assays, and the interdiction of illicit materials. A wide range of science and technology specialties is necessary to accomplish these important activities. The R&D efforts of many scientists and engineers to protect, control, and account for uranium and plutonium are highlighted in this publication.

Measurement is one key specialty that allows us to identify and quantify special nuclear materials. This report describes R&D work in calorimetry, neutron interrogation and measurement, and gamma-ray measurement. Each type of measurement addresses certain aspects of uranium and plutonium assays, and each is useful in a variety of situations. However, because there is no one universal solution to the assay of these materials in every venue, research in these and other measurement methods continues to be refined and strengthened so that users can be confident they are getting the best possible results.

Systems integrate these measurement techniques into a variety of instruments that perform a particular task, such as monitor a portal, or verify that a container holds a declared amount of uranium or plutonium. Often the methods are combined to provide an overall result. For instance, calorimetry measures the total mass of plutonium in a sample once the plutonium isotopic ratios are provided by gamma-ray spectrometry. As an example of an integrated system, ARIES—which combines three nondestructive analysis (NDA) methods—demonstrates the capabilities necessary for weapons-material conversion facilities in both the U.S. and Russia.

Seals and monitoring systems are methods to protect special nuclear materials. Such systems are described in this report which provide both passive and active assurance that special nuclear material in a storage facility is not tampered with or moved.

Measurement data becomes information that is ultimately used to help protect, control, and account for special nuclear materials. Modern electronics, computer systems, and data management are required to ensure that the measurement methods employed by a variety of systems provide reliable and useful information. In this report, several levels of data collection and management are discussed.

Mobile Calorimetry Laboratory

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Much of the nuclear inventory in the U.S. is in a form difficult or impossible to assay accurately using chemistry or most NDA techniques. The problem is most severe for highly enriched uranium (HEU) metal. In many cases, NDA methods for HEU and plutonium do not have suitable standards to provide accurate assays. It has been shown that the calorimetric assay technique (calorimetry plus high-resolution gamma-ray spectroscopy) now used to measure plutonium can be applied to multi-kilogram quantities of HEU. This technique could be used to make secondary HEU as well as plutonium standards for neutron counting and other NDA techniques. The advent of the new secondary standards will lead to reduced and realistic systematic error estimates for the U.S. inventory of special nuclear materials (SNM). Direct assays of items with anomalous contents could also be made with this system.

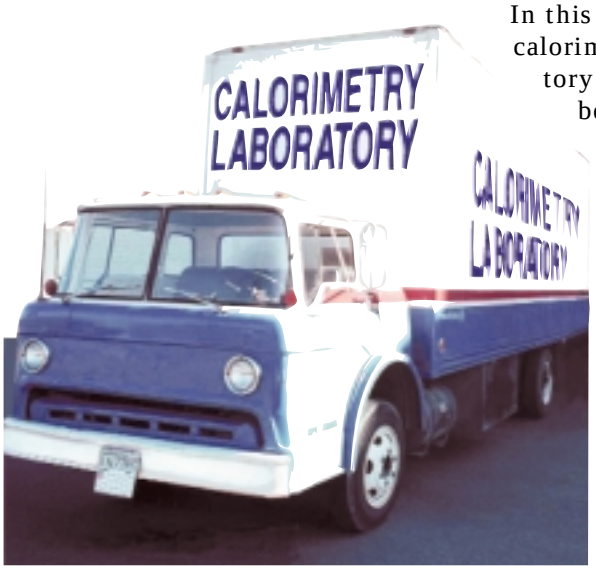

Figure 1. Concept of the Mobile Calorimetry Laboratory.

In this project, a calorimeter laboratory capable of being transported to various DOE facilities will be built. The laboratory will measure the uranium or plutonium content of sealed items selected from various material categories. The measured items will serve as physical standards for other, faster NDA methods. Besides characterizing difficult-to-measure materials, the lab will verify nuclear facility inventory results, solve other assay problems, and help resolve significant inventory differences. The mobile lab will contain at least three calorimeters, capable of measuring the thermal power of SNM in containers, 2, 5, and 13.5 inches in diameter, and one gamma spectroscopy system to determine the isotopic composition. The two smaller calorimeters have already been built from earlier projects funded by the the Office of Safeguards and Security. The largest calorimeter using advanced sensor technology will be built at Los Alamos National Laboratory as part of this project, initiated in FY 2000. The mobile lab will be operated by either DOE personnel or designated contractors.

Milestones

FY 2000: Design trailer and 13.5-inch calorimeter
FY 2001: Construct calorimeter and mobile calorimeter lab
FY 2002: Field-test mobile calorimeter lab.

Calorimetric Assay of Nuclear Materials

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Calorimetric assay is one of the most important accountability techniques for measuring plutonium and tritium. Calorimeters are found in every facility and lab that possess these materials. Up to 28 calorimeters are planned for one DOE facility. Why calorimetry? Nuclear materials release heat as they decay. The amount of heat, or thermal power, generated is directly proportional to the amount of nuclear material in an item. Thermal power is measured very accurately with calorimeters. In addition, thermal-power measurements are independent of the matrix type. There is no way to shield the heat coming from an item. Any added insulation just increases the measurement time; it does not change the final assay answer. Calorimeters are designed to give the same result no matter how the nuclear material is distributed inside the item being measured. Calorimeter measurement precisions of less than 0.1% have been observed in a production facility.

The accuracy of calorimeters in the DOE complex is maintained using small, encapsulated radioactive ^{238}Pu heat standards calibrated in Los Alamos National Laboratory's Heat Standards Lab. Heat standards are certified using special high-accuracy calorimeters with electrical standards traceable to NIST (**Figure 1**). The degree of accuracy of the heat standards is shown in **Figure 2**. Repeat

Figure 1. Heat standard calibration calorimeters.

measurements of the same standards in the Heat Standards Lab over a 30-year period with different analysts and instruments agree to better than 0.01% for powers greater than 0.1 W. DOE started this program approximately 35 years ago at Mound Laboratory; the program was transferred to Los Alamos in 1996.

During the past two years, 85 heat standards and 5 other plutonium assay standards have been calibrated at Los Alamos for 9 DOE facilities and laboratories. In addition to calibration, the Heat Standards Lab supports other DOE facilities in troubleshooting, training, statistical consultation, and standards committees.

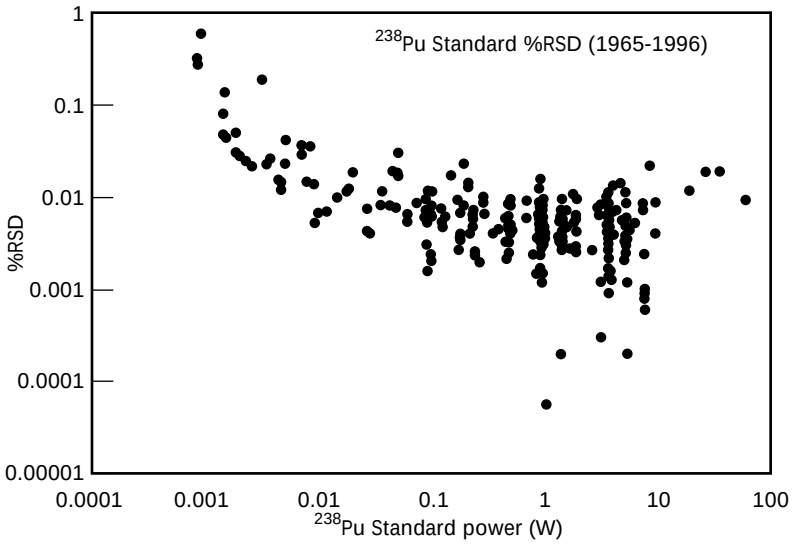

Figure 2. Heat standard accuracies from replicate measurements with Mound Heat Standards Laboratory's calorimeters.

Solid-State Calorimeter

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We designed and fabricated a calorimeter using thermopile sensors. The solid-state calorimeter, fabricated primarily from commercially available components, makes highly precise measurements comparable to those made using much larger, non-portable calorimeters. Using highly sensitive thermopile sensors in calorimetric NDA systems produces more robust calorimeters with excellent portability and a stable baseline output. The data-collection electronics are commercial digital voltmeters; the Los Alamos-developed data-acquisition and instrument-control software, MultiCal, runs under Windows NT® (**Figure 1**).

Figure 1. The highly precise solid-state calorimeter is shown submerged in a temperature-controlled water bath with the electronics and data-acquisition computer on the right.

The footprint of the complete system is 68.6 cm wide by 39.4 cm deep. A water bath provides a constant temperature heat sink, supplying a precise reference temperature stable to ±0.001°C. The total volume of the bath is 36 liters. The current design allows for a dry reference-temperature heat sink. With the water drained from the bath, the calorimeter is easily transported between different locations and is ready to measure within 24 hours (**Figure 2**).

Table 1 shows the relationship between source wattage and measurement precision. Plutonium produces between 2.5 mW/g and 12–20 mW/g for low burnup and high burnup plutonium, respectively. The high burnup range is due to the varying mass fractions of ^{240}Pu and ^{238}Pu. The standard deviation increases slowly with increasing source power, so the standard deviation becomes a smaller percentage as power increases. At a source power of ~10 mW, the relative standard deviation of 6 measurements is 0.11%. The extremely low noise of the heat-flow sensors, less than a microvolt, allows for the high-precision measurements of low-power items. The last column in Table 1 contains the number of replicate measurements made on the source listed in column 1. The zero-power measurements were taken with no heat source within the calorimeter.

Table 1. Solid-State Calorimeter measurement precision as a function of power. Measurements were taken at two different sites over several months.

Power (mW)	Low Burnup Plutonium Equivalent (g)	Average Signal (mV)	Standard Deviation	% Relative Standard Deviation	% Relative Standard Deviation of Mean	Number of Measurements
0	0	4.50×10^{-5}	5.2×10^{-5}	—		7
1.975	0.79	0.02098	4.4×10^{-5}	0.211	0.094	5
4.850	1.940	0.05185	1.1×10^{-4}	0.221	0.090	6
10.716	4.288	0.11393	1.3×10^{-4}	0.113	0.046	6
82.78	33.112	0.8811	1.2×10^{-3}	0.136	0.055	6
423	169.2	4.4833	2.8×10^{-3}	0.0618	0.028	5

The sensor response to heat is linear. A least-squares fit to a plot of calorimeter output (in mV) as a function of source power (in watts) determines the sensitivity (in mV/W) of the calorimeter. The sensitivity converts the voltage signals from unknown items to watts of power. The slope and one sigma uncertainty determined from the zero, 1.98, and 10.7 mW data is 10.6316 ± 0.0042 mV/W.

By the time the project is completed this year, the measurement capability of the solid-state calorimeter will be characterized. The prototype solid-state calorimeter is now available for small plutonium, uranium, or alternative nuclear material items. This calorimeter is ideally suited for making secondary working standards for the calibration of other NDA instruments such as neutron counters.

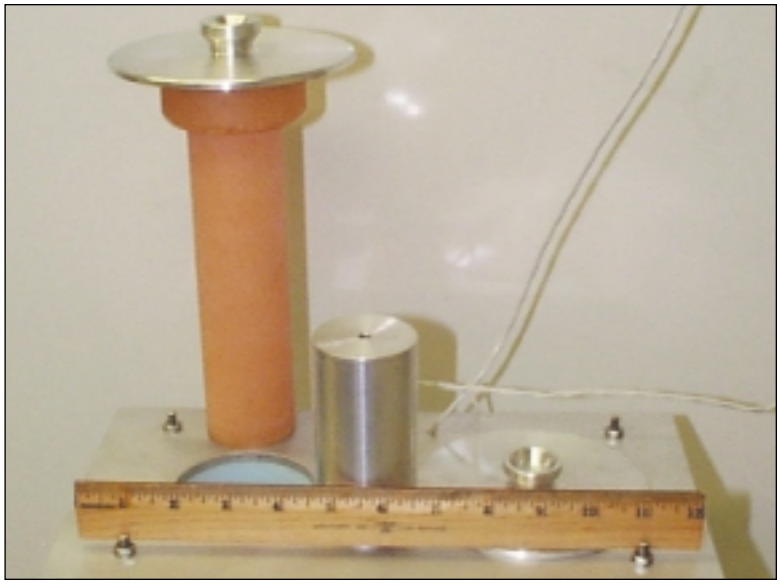

Figure 2. A top view of the calorimeter shows the insulating plug and the item can. The internal dimensions of the item can are 9.65 cm deep and 4.05 cm in diameter. The longer cylindrical tube is the insulating plug and the shorter cylinder is the item can.

Large-Volume Calorimeter Measurements of Enriched Uranium

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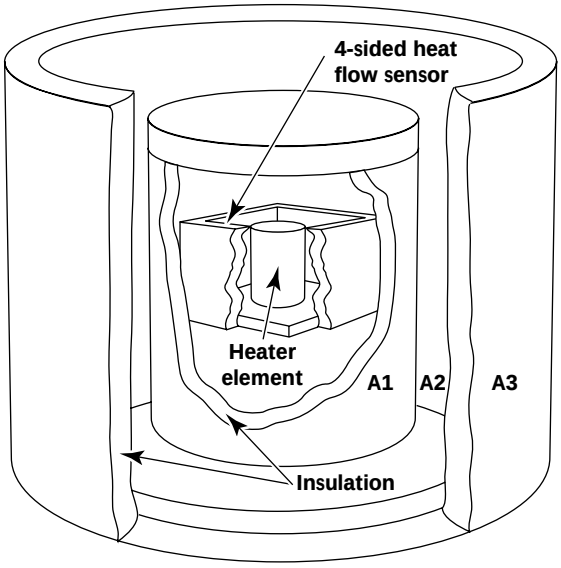
Current assay methods for uranium are expensive or inaccurate. The most accurate technique, destructive chemical analysis, has become almost prohibitively expensive for routine accountability measurements. The other existing technique for bulk-uranium analysis is active neutron assay. Neutron measurements are strongly influenced by the geometry, chemical form of the material, and the matrix composition. This leads to large uncertainties and/or biases.

Calorimetry measurements are inherently independent of the material form and composition. Fabrication of a high-sensitivity, large-volume calorimeter that measures enriched uranium and plutonium in 55-gallon shipping containers will be more accurate and thus provide higher confidence in the special nuclear material (SNM) inventory. Cost savings will also be realized when compared to destructive analysis.

In this project, we will design, fabricate, and test a large calorimeter capable of measuring enriched uranium or plutonium in 55-gallon shipping containers (**Figure 1**). The calorimeter will make it possible to accurately assay the uranium content of objects such as fuel plates, weapons components, and other uranium-process materials. Some material types that could be measured with a large-volume calorimeter include U_3O_8 in poly bottles, metal turnings, skulls, UF_4, metal chips in stainless-steel containers, UO_2 in poly bottles, UAl_x scrap, and tritium or SNM weapons components.

Work has begun on overcoming the technical challenges of scaling a calorimeter to the size of a 55-gallon drum. This is possible by using the same type of solid-state sensors investigated during the development of the solid-state calorimeter. The heat output per gram of highly enriched uranium is 1,000 times less than for low burnup plutonium. The biggest challenge is to develop a non-aqueous reference temperature stable enough to measure highly enriched uranium in large containers. **Figure 2** represents the current test bed for developing a non-aqueous reference temperature. A cascaded system of heaters, insulators, and thermally conductive metals currently control a reference temperature to better than 100 micro-degrees Celsius. With continued funding, a calorimeter capable of measuring 55-gallon drum size containers could be available by FY 2004.

Figure 1. Two 55-gal drums with solid-state, heat-flow sensors on top. Calorimetric assays of drums this size or smaller will be possible at the completion of this project. Current calorimeter capability is limited to items 25-gallon or less with a maximum diameter of 14 inches.



A1 - Inner feedback
A2 - Thermistor 2
A3 - Outer feedback

Figure 2. Schematic of large-volume calorimeter test and development bed.

Fiber-Optic Calorimetry

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Calorimetry combined with gamma-ray spectroscopy is the primary accountability assay technique for plutonium in the DOE complex. Extending this technique to measure highly enriched uranium (HEU) will significantly improve the accuracy of HEU accountability measurements. The technical problem involved in the calorimetric measurement of HEU (93% ^{235}U) is that the thermal power emitted is 1,000 times less than that of ^{239}Pu. Current sensors are not sensitive enough to measure this low power for many forms of HEU.

Fiber-optic sensors are a more sensitive technique to measure the small amounts of heat generated by HEU. The increase in temperature expands a fiber, increasing its length and changing its index of refraction. This changes the total number of wavelengths of light traveling through the fiber as the temperature rises. For example, a one-degree change in temperature in a one-meter length of silica fiber causes the number of wavelengths to change by about 100 radians. Currently, commercially available hardware is can measure wavelength changes of 10^{-6} radians. Thus, the potential sensitivity of one-meter fiber is about 10^{-8} °C, orders of magnitude more sensitive than current technologies.

The calorimeter is set up in a Michelson interferometer configuration so that the wavelength shifts in a sensor arm are measured relative to a reference arm. A schematic of the measurement setup is shown in **Figure 1**. A 5-inch-diameter, fiber-optic calorimeter using a laser operating at 1310 nm is now being tested.

Two fiber-optic calorimeters have been built and tested using electrical heaters to supply thermal power. The 1-inch-diameter calorimeter had a baseline sensitivity of 0.2 radians, corresponding to 3 mW of thermal power, corresponding to 1 milligram of Pu. **Figure 2** is a photo of the calorimeter thermals. A 5-inch-diameter calorimeter, capable of measuring a 3-liter size sample, showed at best a sensitivity about a factor of 2 better than the best traditional calorimeter. Research is continuing on more stable fiber-optic sensors, cutting out noise due to item insertion, and a more quiescent temperature control system.

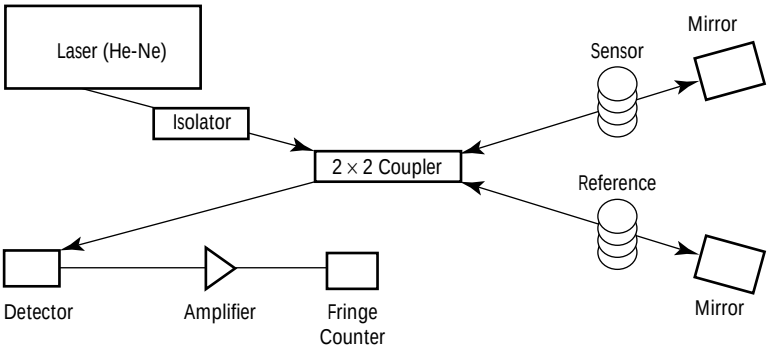

Figure 1. Fiber-optic calorimeter.

Figure 2. 1-inch-diameter, fiber-optic calorimeter.

A Robust Prediction Algorithm for Faster Calorimeter Assays

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Calorimetry is an essential component of nuclear-weapons measurements. In some cases, it is the only measurement that achieves the required precision and accuracy. Unfortunately, for a large number of samples, the assay time is very slow—on the order 4 to 24 hours—and calorimetry becomes the limiting factor for material disposition. The long assay times for some samples depend on the heat flow through the sample, not on the calorimeter. Therefore, the assay time cannot be improved by improving the calorimeter's design.

One approach to address this limitation is prediction. Prediction improves the assay time by removing the requirement that the calorimeter and item come to equilibrium prior to completing the assay. Measurement times should be reduced by a factor of about 1.5 to 3. Reduced measurement time decreases the need to buy additional calorimeters to achieve the same level of throughput.

Very little work has been done on calorimetry prediction and the methods used have been somewhat simplistic. For example, the original Mound algorithm was a 3-point fit to a single exponential.

A more effective approach uses all of the acquired data and employs more modern function-minimization methods. When all of the calorimeter data is fit, including the initial transients generated after the insertion of the calorimeter can, typically 5 to 7 exponential terms are required to fit the data. This approach has been incorporated into the multi-exponential prediction algorithm (MEPA), which has been applied to over 600 calorimeter data sets. Data can be reliably fit down to the level of the calorimeter noise.

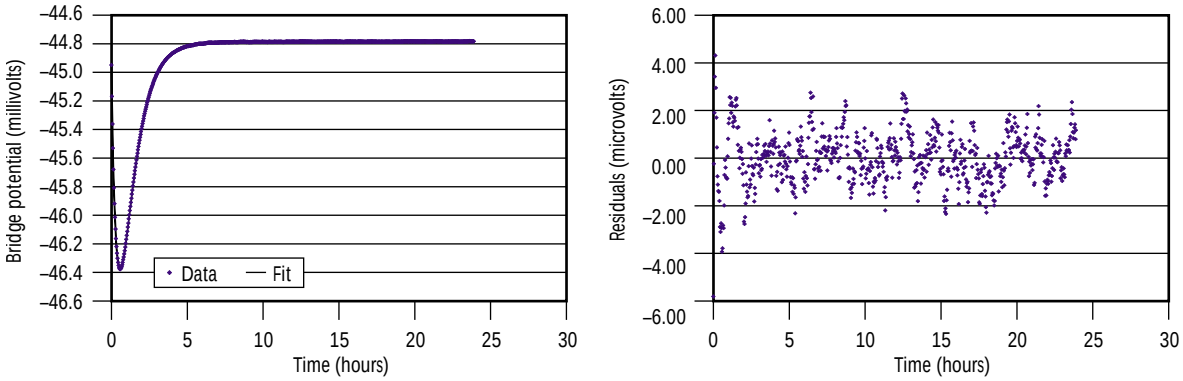

Figure 1. (left) Bridge output of a 5-inch-diameter calorimeter measuring 1 kg of HEU oxide. The open circles are the raw bridge voltages and the solid line is a best fit to the data using 6 exponential terms. (right) Residuals of the fit, raw data minus fit value, as a function of time. The magnitude of the fluctuation, 2–4 microwatts, is comparable to the noise in the raw data.

The effectiveness of the MEPA appears to be good. Predictions made at about 60% of the equilibrium time are typically within 10 microvolts and often less than 1 microvolt of the equilibrium value determined by averaging raw data at equilibrium. This is good evidence that MEPA is robust and accurate considering predictions were made to over 600 data sets from 7 different calorimeters, and the one standard deviation noise of the calorimeters ranged from several microvolts to tens of microvolts. An example of the good fit achieved using MEPA is presented in **Figure 1**. The solid line is a fit to the data with a functional form that is the summation of 6 exponential terms, plus a constant. The constant is the equilibrium value of the bridge potential. The mean of the data at equilibrium is 4.478334 millivolts with a standard deviation of 0.2658 microvolts. The constant of the fit was found to be 4.478378 millivolts matching the average of the data within one sigma.

The MEPA will be incorporated into the data-acquisition software, MultiCal (see page 14). The addition to the data-acquisition software will be NQA1 certified and available to users at the beginning of FY 2002.

MultiCal Calorimetry Software

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MultiCal, software developed by Los Alamos National Laboratory, operates multiple calorimeters from a single computer. The fast software offers more transparent procedures for calorimetry measurements. MultiCal is written in C++ under the Windows NT operating system. Working within the framework of object-oriented design, the software incorporates an integrated module process technique. MultiCal controls all device I/O and data acquisition and analysis. The software provides equilibration detection, predictive capability, and servo control modes of operation. GPIB and serial communication protocols are supported by the software with ethernet capability to be added later.

MultiCal's handling of different calorimeter configurations is flexible. The software interfaces to digital voltmeters and controls a variety of meters. It features a set of on-screen forms where the user can specify measurement parameters and select options and access the on-line help menu. Measurement options include Baseline, Assay, and Basepower. MultiCal provides functions to control the calorimeter instrument hardware and compute results using the data acquired during the requested operation. After the result in Watts is generated, the data is displayed on the video monitor. Numerous internal diagnostics automatically verify communication paths, detect instrument and hardware faults, and test on-line data consistency. All of the raw data is written, along with the measurement results, to a Microsoft Access database. Environmental room-monitor data are collected via a serial digital voltmeter and the data from this meter are also stored in a table in the database. Results are available in ASCII form in files that can be accessed by commercial spreadsheet products. MultiCal has been interfaced to another computer system and is capable of receiving commands to start and stop measurements over a serial interface from the auxiliary computer system (**Figure 1**).

The current version of MultiCal has been installed at several DOE facilities and is operating 25 calorimeters. MultiCal has been tested and validated at the Rocky Flats facility and passed all tests. The software has received NQA-1 certification.

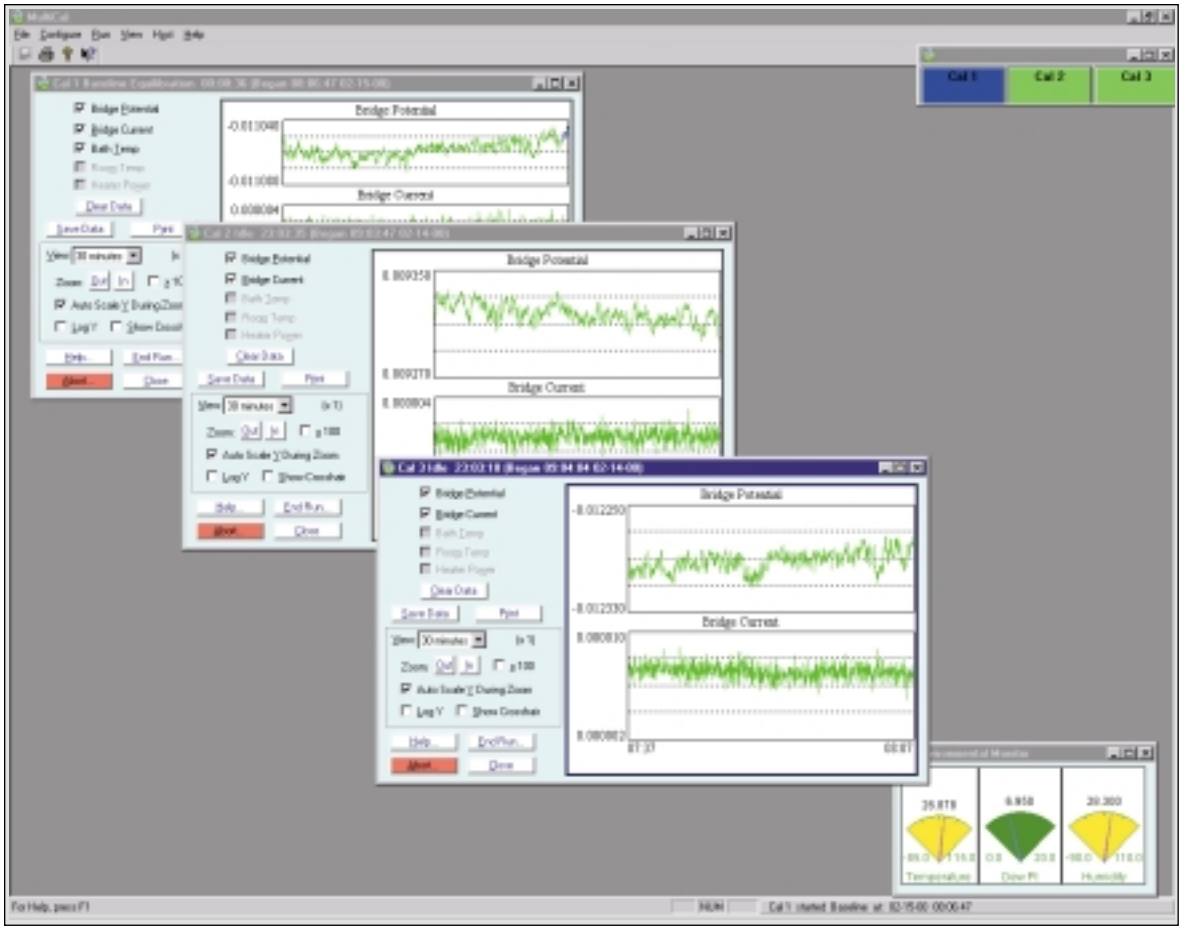

Figure 1. MultiCal main screen displaying the operation of three calorimeters with active data collection, the calorimeter status information, and the environmental monitor readout.

Advanced Epithermal Neutron Multiplicity Counter

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With support from Los Alamos-directed research programs and capital-equipment funds from the Office of Safeguards and Security, we built and tested a new Epithermal Neutron Multiplicity Counter (ENMC) to reduce errors and the time required to perform quantitative, neutron-based NDA measurements of bulk items. We are also completing a new inventory sample counter (INVS) insert for small samples, taken routinely for destructive analysis.

Application of neutron-based NDA methods for materials control and accounting (MC&A) is limited by precision or the counting time for many materials, especially plutonium-bearing residues containing unknown impurities. Reducing the counting time offers many advantages to the MC&A community by saving costs, reducing radiation exposure, and reducing the potential for diversion. Improving the precision also increases the quality of MC&A data.

Monte Carlo simulations and laboratory tests have shown that by increasing the fill pressure of ^{3}He proportional counters and by reducing the moderator, a high-counting efficiency can be achieved along with a low die-away time. The ENMC has an absolute efficiency of 65% and a die-away time of only 21 μs. The ENMC is designed for measurements of items in DOE-standard 3013 containers. The INVS insert increases efficiency to 80% with the same die-away time (**Figure 1**). The ENMC–INVS is designed for small samples, such as those typically collected for destructive analyses. The ENMC and ENMC–INVS

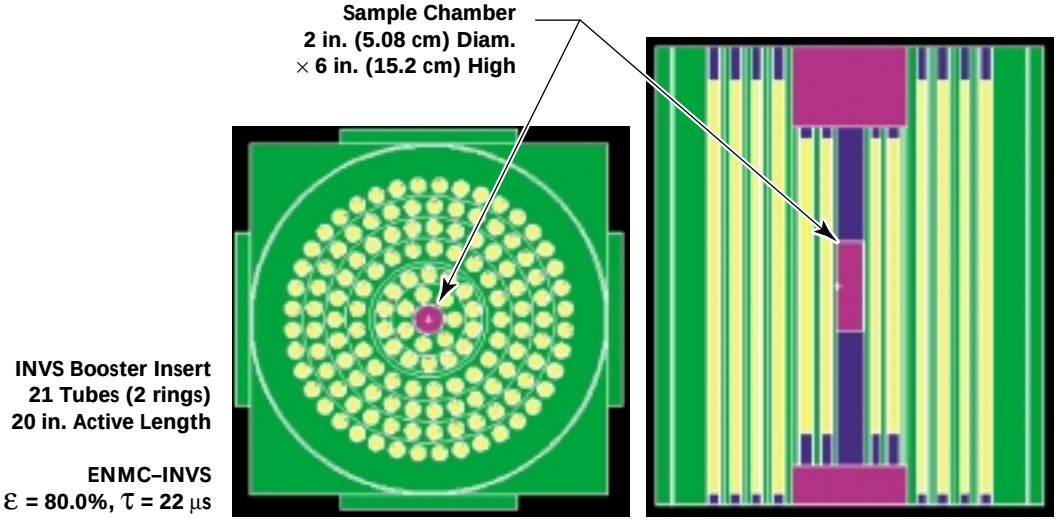

Figure 1. The ENMC–INVS design includes small graphite end plugs and a removable sample chamber.

have performance characteristics superior to all existing neutron counters and are competitive with calorimetry. Compared with present state-of-the-art thermal neutron multiplicity counters (thermal NMCs), "figure-of-merit" calculations and recent measurements show that measurement times are reduced by a factor of 5–20 for a wide range of plutonium masses and impurity levels. The improvement factor is greater for high-impurity levels than for pure materials.

In parallel, we have also designed and tested new electronics circuits using an experimental neutron multiplicity shift register (EMSR) that improves assay precision for active and passive neutron coincidence and multiplicity assays (**Figure 2**). The improvement here is approximately a factor of 2 in counting time. Combining the ENMC and multiplicity shift register technology yields a system faster by factors of 10–40 compared with current thermal NMCs.

This project will evaluate the ENMC, the ENMC–INVS, and the EMSR in DOE facilities. We will also develop an active-mode ENMC for highly enriched uranium. In the active/passive mode, we will evaluate the system for the assay of neptunium and neptunium–plutonium mixtures. This technology will be transferred to the private sector in FY 2001. This technology builds on existing product lines and can be commercialized with a minimum of development. We estimate an epithermal NMC system will be only ~20–25% more expensive than a typical thermal NMC system.

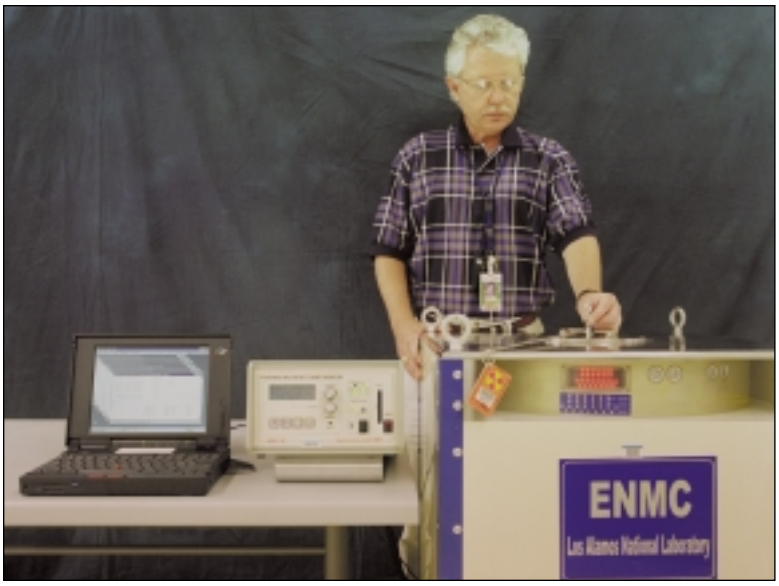

Figure 2. The Epithermal Neutron Multiplicity Counter and experimental Multiplicity Shift Register system in operation.

Fast-Neutron Counters To Rapidly Verify Bulk Uranium

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Assaying bulk uranium in an item is difficult because uranium is a weak emitter of spontaneous-fission neutrons—the penetrating radiation signal that determines the mass. In fact, so few neutrons are emitted that a passive measurement is not practical for most uranium items. Active measurements overcome this obstacle.

In an active measurement, an external neutron source induces fission events in the item. In active coincidence counting, the coincidence neutrons from the induced fissions provide a quantitative measure of the amount of material present. A major limitation with this approach is that the interrogating neutrons interfere with the induced coincidence signal and so reduce the precision of the measurement. For this reason, the Active Well Coincidence Counter (AWCC) technology requires long assay times to achieve the necessary precision for the nondestructive assay of bulk uranium.

This problem is minimized by using coincidence counters with a smaller neutron die-away time, the mean life-time for detecting the induced fission neutrons. Fast-neutron counters with shorter die-away times enable greater throughput, saving costs and reducing radiation exposure to personnel. Measurements of all types of uranium, including HEU metal, bulk oxides, residues, and waste are possible. Also, assays of materials such as unmeasured inventory, excess defense materials, and various process materials are improved and substantially speeded up.

Several options are available for building an assay system that provides a large gain in measurement sensitivity and measurement throughput over current counters. The different detection systems include solid-state scintillator detectors, high-pressure ^{3}He detectors, and liquid scintillators. Solid-state detectors (for example, BC454/BGO phoswich detectors) have a small die-away time but require sophisticated instruments. High-pressure ^{3}He tubes offer slightly longer die-away times but are very simple to operate. Liquid scintillators offer two orders of magnitude reduction in neutron die-away time but are more complex to operate.

We built a prototype Fast-Neutron Coincidence Counter (FNCC) using solid-state scintillator detectors (**Figure 1**). The BC454/BGO phoswich detectors have advantages over traditional detectors because their die-away time of about 4 microseconds results in less interference from the interrogating source. Also, a unique feature of these detectors is that the neutron energy information can discriminate between induced neutrons and those arising from the interrogating source, removing the latter counts. The FNCC design using phoswich detectors can be optimized to outperform the current AWCC. However, this approach has a low neutron detection efficiency, which negates some of the gains achieved by the small die-away time.

We have also adapted the existing, passive Epithermal Neutron Multiplicity Counter (ENMC) for active uranium assay. The ENMC uses new technology, namely 10-atmosphere ^{3}He tubes, to provide a very high efficiency counter. The die-away time of 20 microseconds is not as short as the FNCC's, but the higher efficiency compensates for this. We have modified the ENMC for active uranium

assay and completed an initial calibration using uranium standards. Additional calibration and inventory measurement activities will be carried out this year to evaluate this faster approach with the goal of verifying hard-to-measure inventory items. We expect the ENMC to reduce uranium count times by a factor of 5 to 10 over the AWCC. Shown in **Figure 2**, is the expected precision for a 1,000-second count time for the three different types of active counters. These results were obtained from a figure-of-merit code that predicts neutron-counter performance. The ENMC outperforms the AWCC, and also the FNCC at higher uranium masses.

Figure 1. The Fast-Neutron Coincidence Counter uses BC454/BGO phoswich detectors to quickly detect neutrons, thereby reducing the interference from interrogating neutron sources.

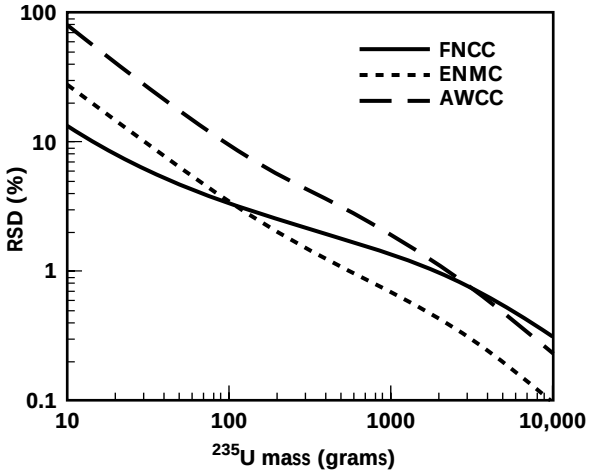

Figure 2. Comparison of the performance of three active neutron counters. The precision is shown for a 1,000-second count time over a mass range of 10 to 10,000 grams of ^{235}U.

Neutron-Capture Counter for Fast-Coincidence Assay of Plutonium in Residues

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We carried out computational and experimental evaluations of commercially manufactured neutron detector elements: (1) variable-diameter, boron-loaded scintillating plastic fibers and ribbons and (2) ^{6}Li/scintillator-clad fiber ribbon. Neutron-coincidence well counters were also studied based on both types of detector elements. We wanted to compare computational and experimental results for a new detector that uses a stack of thin layers of neutron-capturing scintillator with plastic fibers. This will create an efficient, short-die-away-time neutron detector that also discriminates against gamma rays.

The well-counter design studies determined whether the commercially manufactured detector elements can theoretically achieve the neutron detection efficiency (ε) and die-away time (τ) necessary for the new generation of neutron multiplicity counters. Such counters will be essential for assaying plutonium scrap in residue forms because of the high, uncorrelated neutron yields that destroy the detection sensitivity of conventional neutron-coincidence counters. Conventional neutron-coincidence counters are based on polyethylene-moderated ^{3}He detectors whose neutron capture medium (^{3}He) is physically separate (as a gas) from a solid polyethylene moderator (^{1}H). Shorter neutron die-away times—achieved by homogenizing concentrated neutron-capture target nuclides (^{10}B or ^{6}Li) in a moderator (^{1}H) medium (such as plastic fiber and/or scintillator binder)—permit shorter coincidence gate widths, reducing the accidental coincidence rate proportionally.

The experimental evaluations strongly favor the ^{6}Li/scintillator-clad fiber ribbon over the boron-loaded scintillating plastic fiber in its ability to discriminate against gamma rays. The well-counter design studies have demonstrated that practical assemblies of both types of experimental detector elements can achieve the required neutron detection efficiency (ε = 50%, comparable to the counters based on ^{3}He proportional detectors). They also reveal that the die-away times of these counters are 1/5 to 1/20 those of the polyethylene-moderated ^{3}He proportional detectors, and that the shortest of these (τ = 3-5 μs) is achieved with the ^{6}Li/scintillator-clad fiber ribbon.

Our prototype well counter based on the ^{6}Li/scintillator-clad fiber ribbon is shown conceptually in **Figures 1** and **2**. Figure 1 describes the individual detector element and the configuration of four detector elements in a well counter. Figure 2 illustrates the assembly of a single element and the arrangement of the four elements with the photodetectors in place.

We tested the new counter in the laboratory with a small detector element consisting of 20 layers of ^{6}Li/scintillator-clad fiber ribbon. The results of these tests refined the design of the well counter and are also being compared to ongoing computational simulations. Fabrication of the commercial prototype well counter has begun. Delivery of the first element is imminent.

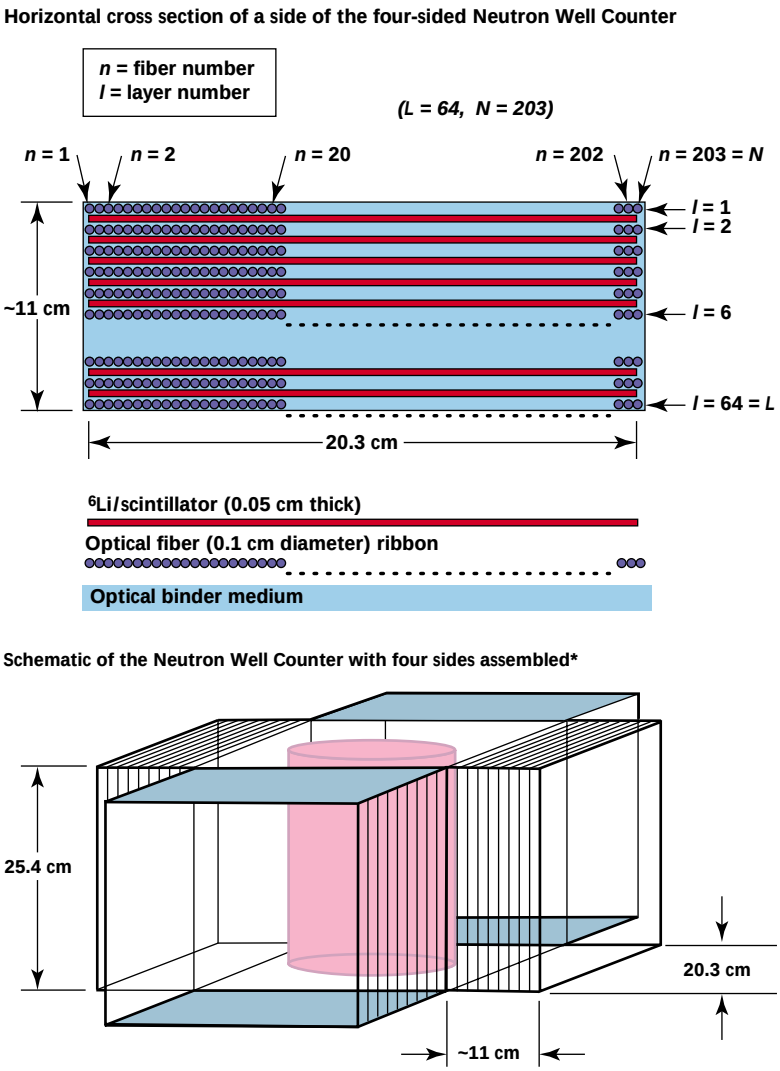

Figure 1. (top) The neutron-detection portion of one detector element is illustrated in a cross-sectional view of a stack of fiber ribbon/^{6}Li scintillator layers. Each side of the prototype four–sided neutron well counter will consist of a stack of alternating fiber ribbon and ^{6}Li-scintillator sheets, 64 layers of each, in a hydrogenous optical binder medium. The fibers in this slab will be oriented vertically and coupled to photomultiplier tubes at the top and bottom. Each ribbon, consisting of 1-mm-diameter, wavelength-shifting optical fibers, is 20 cm in width and 0.1 cm thick. (bottom) The neutron-detection portions of the four-sided neutron well counter are arranged as shown. The red object represents an item placed in position for counting coincident neutrons.

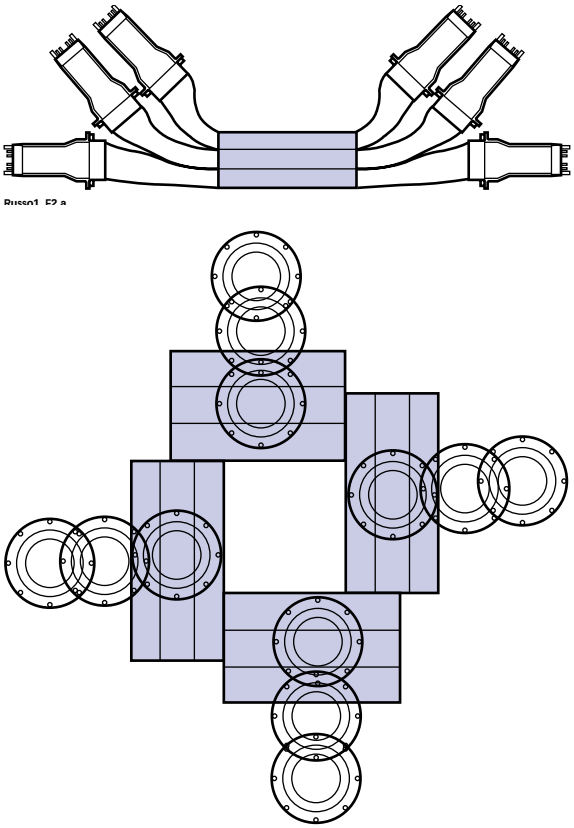

Figure 2. Six photomultiplier tubes (three at each end) are coupled to each of four 64-layer elements. The shaded area represents the horizontally oriented fibers in the three 21-ribbon layers of the active portion of one 64-layer element. The overall length of the active portion is 25 cm, and its thickness is ~11 cm (horizontal and vertical dimensions, respectively, of the shaded region (top). A plan view of the four elements oriented vertically in the well-counter arrangement is shown in the bottom drawing. The cross sections of three 21-ribbon layers are shown for each of four elements. The overall width of each element (long dimension of each shaded region in (bottom) is 20 cm. The well dimension for the arrangement illustrated can be varied from 10 to 20 cm. The sketch is a modified version of a drawing provided by the Bicron Corporation.

Neutron Coincidence Counter

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International treaty verification and U.S. nuclear material control and accounting (MC&A) require the capability to determine the mass of plutonium present in an assembly, container, or similar object. This project is developing a highly portable and efficient means of determining the plutonium mass.

For treaty verification and MC&A, the variety of samples ranges from relatively simple plutonium metal stored in barrels, to waste streams in which plutonium oxides, plutonium fluorides, as well as plutonium metal may be present (mixtures), to full nuclear assemblies whose materials could potentially interfere with the measurements.

We are developing a gamma-ray and neutron detector hybrid that will determine the unknown mass and isotopes of plutonium in a variety of containers and configurations. The neutron detectors are ^{6}Li glass scintillators configured to measure individual neutrons and neutrons in coincidence. The gamma-ray detector is a high-purity germanium detector. From the neutron measurements, we will obtain an effective mass of ^{240}Pu, the primary neutron emitter in plutonium. From the gamma-ray detector, we will extract the plutonium isotopes and combine these results with the neutron measurements to determine the actual mass of plutonium present.

The number of coincidences between the two ^{6}Li detectors is directly related to the mass and isotopes of the plutonium inside the container. The methodology also allows us to remove thermalization (slowing of the neutrons) effects resulting from materials being present between the plutonium and

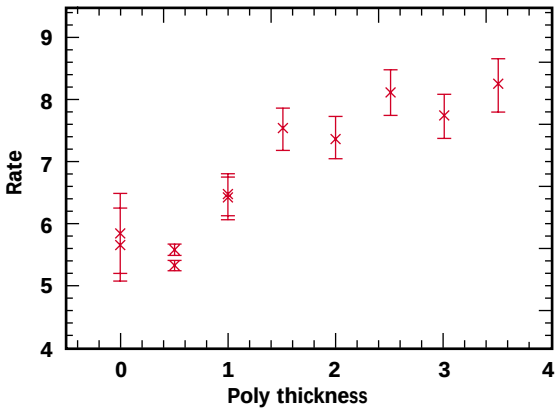

Figure 1. The dependence of the ratio of singles neutron counts to coincidence neutron counts plotted as a function of polyethylene thickness.

the neutron detectors. A ratio of measurements by two neutron detectors is used so that effects of measurement geometry are removed and the measurement becomes independent of configuration. **Figure 1** illustrates this. The increase of the neutrons detected in each detector and the coincidence rate is plotted as a function of polyethylene thickness (analogous to a absorber found in weapons configurations). This allows us to determine the amount of thermalization that has occurred and remove the effects from this thermalization on the measurement.

We are planning to reduce the size of the detection system to make it truly portable. **Figure 2** shows the present size and the proposed size. This system is still being tested, but we expect that within the next year to achieve the capability to determine the mass of plutonium in unknown samples.

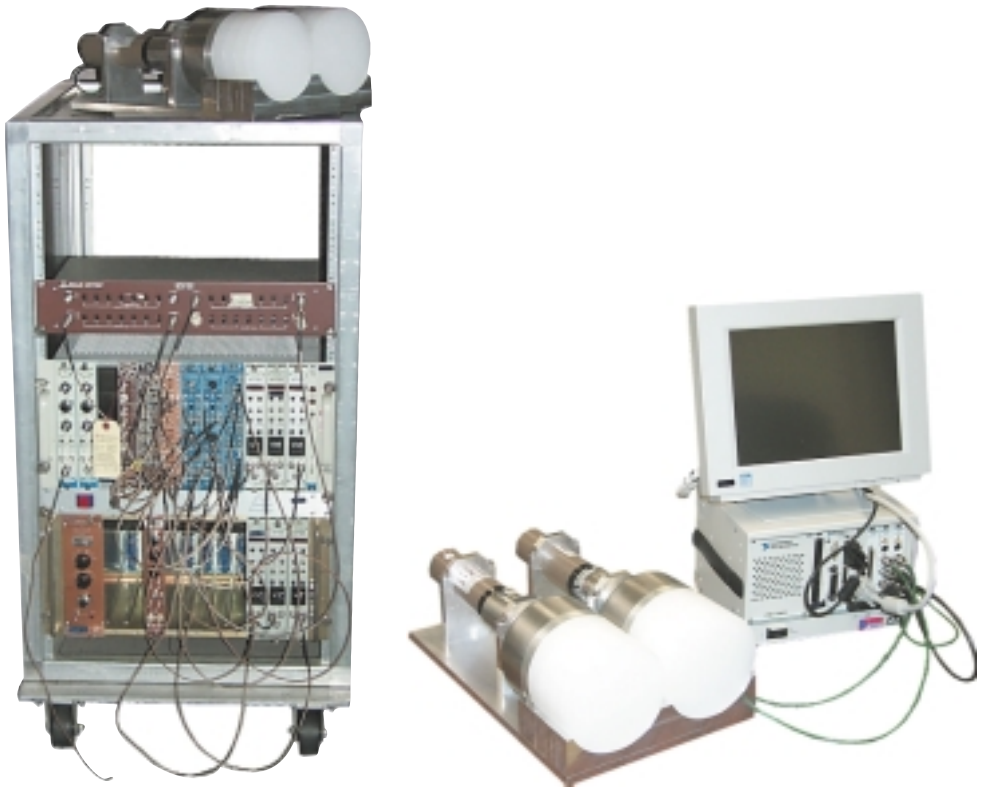

Figure 2. Current and future design of the neutron coincidence portion of the system (relative scale).

International Neutron Coincidence Counting Software

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Neutron-coincidence counting software is used with thermal-neutron coincidence counters for passive and active NDA of plutonium and uranium, respectively. Passive counting is based on measuring correlated fission neutrons spontaneously emitted by the plutonium. Active counting is based on measuring correlated neutrons from fissions induced in uranium by external sources. The neutron detectors are usually ^{3}He proportional counters embedded in polyethylene. Both passive and active methods use time-correlation electronics known as shift-register coincidence circuits to determine the fission rates and thereby the assay masses. The neutron coincidence counting software collects data from the shift-register circuits and processes the data using algorithms appropriate for the type of assayed material. Many passive and active analysis methods are built into the software.

The coincidence-counting program INCC (International Neutron Coincidence Counting) is presently used at numerous facilities throughout the world. It was derived from the older NCC code to include features needed by the International Atomic Energy Agency. A companion code called DEMING works with the INCC code to produce calibration curves from calibration data. **Figure 1** shows the main menu of the INCC code and two submenus for illustration (the calibration

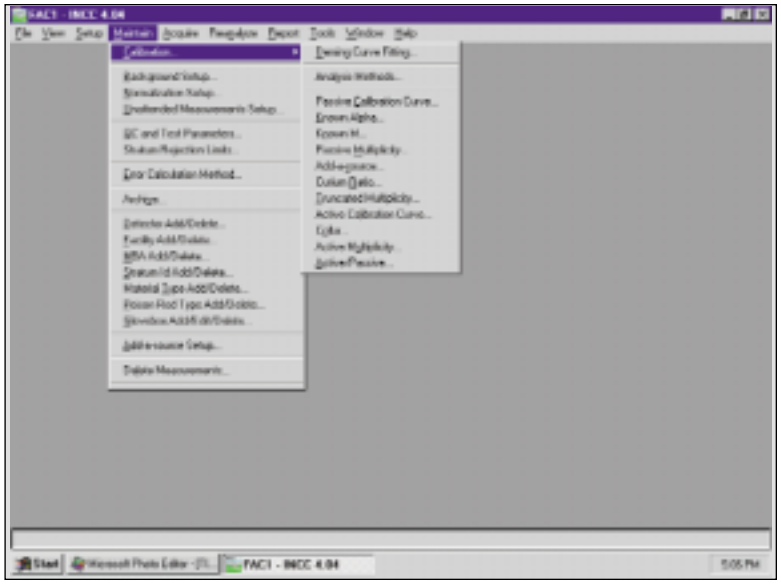

Figure 1. The main menu of the INCC code showing two submenus for maintenance operations. The calibration submenu accesses dialog boxes to enter calibration parameters for various assay techniques.

submenu item in this example). The menu shown under calibration provides access to the DEMING code and to dialog boxes for entering the calibration parameters used by the various analysis methods.

Figure 2 shows the dialog box used by the operator in preparing for an assay or verification measurement. After the operator enters the sample data in this box and clicks OK, the measurement and analysis are performed automatically to produce the assay mass and error.

Figure 3 shows the main window of the DEMING code. In this example, two sets of plutonium calibration data points produced two calibration curves by least-squares fitting—one curve corrected for neutron multiplication (linear) and one uncorrected (nonlinear). The multiplication-corrected rates are much less sensitive to geometric and density variations in the plutonium samples and therefore show less scatter than the uncorrected rates. The dashed lines show the 95% confidence bands for the two curves.

New versions of the INCC code are occasionally introduced to accommodate new analysis methods. Custom versions of INCC have been produced for applications too specialized for general use worldwide. A Russian version of INCC will be available soon.

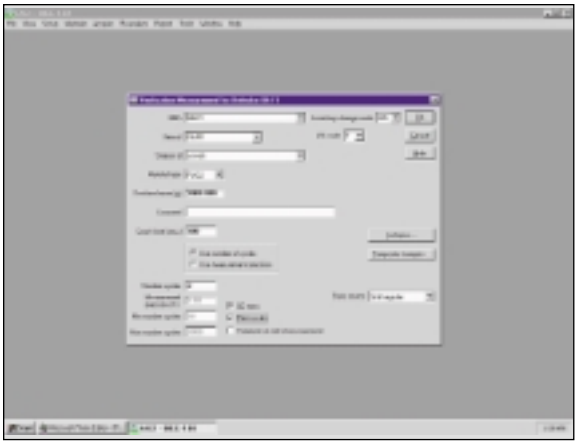

Figure 2. The verification dialog box in the INCC code showing the data selected and entered by the operator to initiate an assay or verification measurement.

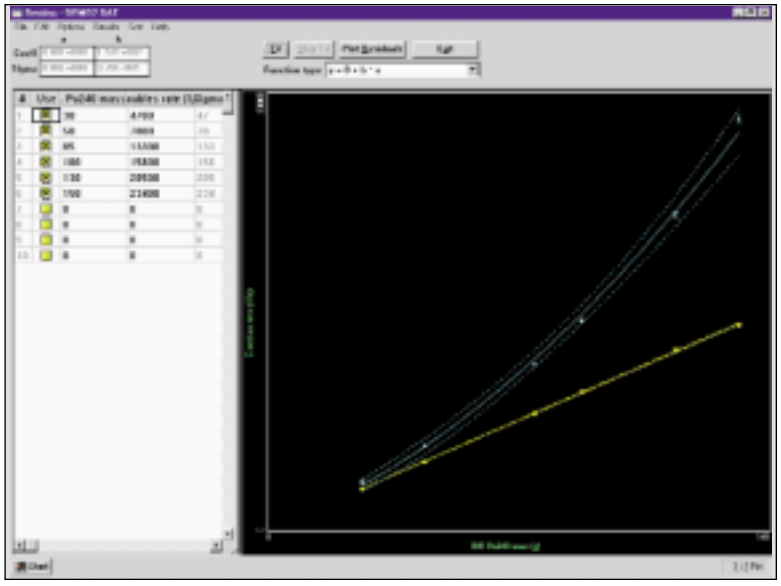

Figure 3. A display from the DEMING program showing the results of least-squares fitting to produce two calibration curves: one for doubles (coincidence) rate vs. effective ^{240}Pu mass and the other for multiplication-corrected doubles rate vs. effective ^{240}Pu mass. The 95% confidence bands are also shown. Note the much tighter confidence band for the multiplication-corrected curve.

Active Interrogation Package Monitor

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Preventing the illicit movement of special nuclear material (SNM) is still the most effective nonproliferation barrier. Although pedestrian and vehicle portal monitors prevent the movement of SNM across the boundaries of nuclear facility, these passive monitors are less effective at airports and border crossings. Passive monitoring methods are not completely adequate for searching luggage and packages for shielded SNM. This has resulted in the need for new technologies to monitor and search for SNM.

Figure 1. The Active Interrogation Package Monitor can detect the presence of SNM in a few seconds even when the SNM is heavily shielded.

One technique being developed at the Los Alamos National Laboratory is using active neutron interrogation to rapidly search containers, packages, and luggage for hidden SNM. Active neutron interrogation has been used for many years for assaying the quantity of SNM in waste containers and 55-gallon drums. This technique has also shown promise for assaying SNM in the presence of other radioactive materials. Neutron interrogation is so sensitive that it can reveal the presence of the SNM in a couple of seconds, even when the SNM is so heavily shielded that no passive signature is present. A prototype device based on this technology has been developed and is referred to as the Active Interrogation Package Monitor (**Figure 1**).

In the Active Interrogation Package Monitor, packages are interrogated with neutrons produced by a pulsed-neutron generator. This generator is a small, electrically powered accelerator. As a result, there is no measurable radiation when the package monitor is turned off. In addition, the interrogating flux of neutrons is sufficiently small so that radiation exposure is not a concern for the package being searched.

Within the walls of the package monitor are multiple detectors that look for neutrons resulting from interactions between the interrogating flux and any SNM that may be present. The package monitor searches packages up to one cubic meter. It can interrogate a high rate of packages because the interrogation time is only a few seconds.

The interrogation technique can be completely automated. The computer interface and operational controls are designed to make the system very easy to use. In addition to making the package monitor safe and easy to operate, we are working to make it compact, portable, and economical.

The prototype has undergone extensive experimental evaluation with excellent results. Design modifications of the package monitor are being pursued to reduce external dose rates to levels acceptable in a public area such as airport x-ray machines.

Active neutron interrogation represents a safeguard technology that will improve the ability to rapidly monitor for SNM and aid in preventing the smuggling of SNM. Although the package monitor was not designed to search every piece of luggage that passes through every airport or across every border, it should be a very useful nonproliferation tool. The unit is currently being prepared for field tests with the U.S. Department of Customs.

Room-Temperature Semiconductors To Determine HEU Holdup and Uranium Isotopics

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Room-temperature, semiconductor radiation detectors are small, portable, and inexpensive but are hindered by poor energy resolution. Energy resolution is what makes the identification of one radioactive isotope from another possible. This poor energy resolution has limited the utility of room-temperature detectors (**Figure 1**). This project aims to eliminate this limitation and develop a portable tool applicable to Material Control and Accounting (MC&A) as well as other safeguards areas.

Figure 1. Typical room-temperature gamma-ray detectors are highly portable but are limited by poor ability to differentiate between the many different gamma-ray emitting radionuclides.

We are developing a technique to dramatically improve the energy resolution of cadmium-zinc-telluride (CdZnTe) detectors with no loss in efficiency by using pulse-shape analysis. Because this technique relies on digital signal processing, it will be far less expensive than the sophisticated (and hard to manufacture) mechanical methods now being employed to improve energy resolution in CdZnTe such as co-planar grid techniques.

The poor energy resolution of CdZnTe is due to incomplete charge collection, usually referred to as ballistic deficit. The energy deposited is only partially collected and the fraction collected depends on the position of deposition within the detector itself (the amount of ballistic deficit depends on the depth in the crystal at which the interaction occurs). Our technique for correcting this problem is to find the interaction location by analyzing the charge pulse from each detected gamma ray to determine the depth of the interaction(s) in the crystal. This allows us to correct for the ballistic deficit and determine the actual energy deposited. **Figure 2** illustrates this process and shows the anticipated enhancement compared to traditional CdZnTe as well as compared to the well-known, liquid-nitrogen-consuming and expensive germanium-based detectors (HPGe).

Figure 3 illustrates how the ballistic deficit depends on the interaction location in the detector. In general, the slope of the linear part of the pulse is proportional to energy and the rise time is proportional to depth. However, our pulse-shape analysis uses a much more sophisticated approach than simple rise-time sorting or other parametric methods. All the information in each pulse is extracted and used to determine the

interaction depth. Each depth will have an associated "correction factor" for ballistic deficit. This method will work for events consisting of single or multiple interactions.

The pulse-shape analysis (patent pending) was developed for another application with HPGe detectors, but it seamlessly translates to our application. This type of pulse-shape analysis could be extended to any room-temperature semiconductor material. Pulse-shape analysis will increase the number of potential semiconductor materials suitable for spectroscopic applications because of the dramatic improvements that can be achieved.

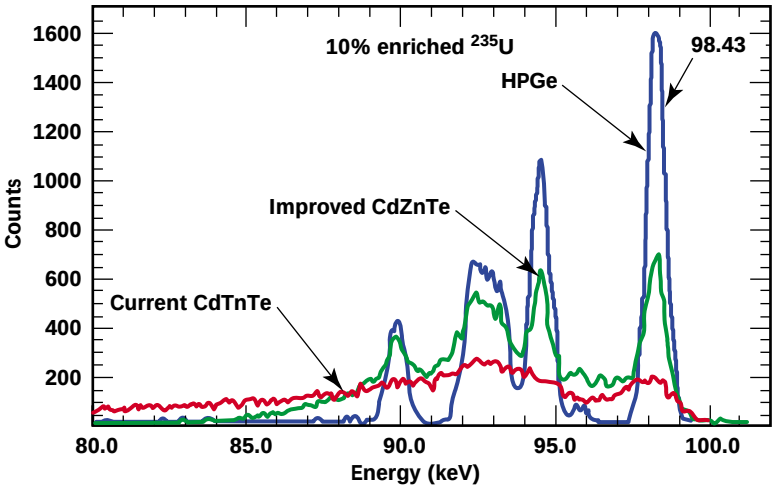

Figure 2. Comparison of ^{235}U spectra from a High-Purity Germanium (HPGe) detector (blue), present CdZnTe detector technology (red), and predicted digital enhancement of CdZnTe (green). The lack of peaks in the red line illustrates the limited usefulness of present CdZnTe detector technology as insufficient detail is available to unambiguously identify this material as enriched ^{235}U.

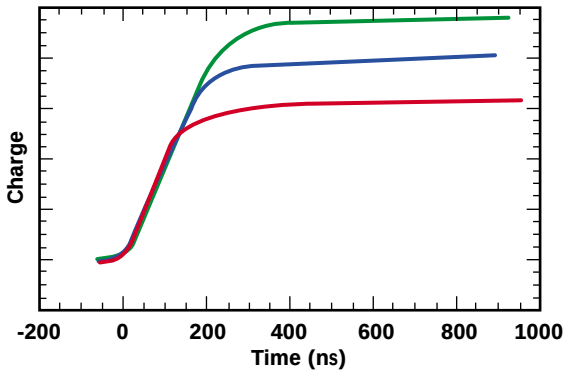

Figure 3. These three charge pulses represent the same energy deposition at three different depths in the crystal. The difference in the maximum charge illustrates the ballistic deficit problem with this material; however, we will use this information to correct the collected data.

Development of CdZnTe Detectors for Domestic and International Safeguards

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Cadmium zinc telluride (CdZnTe)—a new, wide-bandgap semiconductor—has numerous applications in nuclear-material safeguards and nonproliferation. Because of its high resistivity, CdZnTe is ideally suited for gamma-ray detection at room temperature. CdZnTe also has a high atomic number and density, attributes essential to gamma-ray spectroscopy. Methods have been developed to overcome size limitations of compound semiconductors, enabling the development of detectors capable of accurately measuring the energy deposited by gamma rays with very high detection efficiency. Compact, hand-held CdZnTe detectors have been developed that have significantly better pulse-height resolution than scintillation detectors and are easier to use than high-purity germanium detectors in applications that require portability. The counting efficiency is beginning to approach that of NaI(Tl) detectors currently used in safeguards applications.

CdZnTe resolves gamma rays emitted by special nuclear materials, enabling far more accurate measurements of fissile isotopes than previously possible with NaI(Tl). For comparison, pulse-height spectra measured by NaI(Tl) and CdZnTe detectors are shown in **Figure 1** for low-burnup plutonium. Improved resolution is needed in cases where samples are contaminated with isotopes that emit gamma rays which interfere with those emitted by fissile materials. All these applications can benefit from the use of CdZnTe: in situ verification of plutonium isotopics or uranium enrichment (International Atomic Energy Agency); holdup measurements of ^{235}U in the presence of $^{232}U/^{232}Th$ (9206 Facility at Y-12); the measurement of ^{235}U or ^{237}Np in the presence of plutonium (Los Alamos' Plutonium Facility and Rocky Flats); and the measurement of fission products in spent fuel (Idaho National Engineering and Environmenal Laboratory).

Los Alamos' safeguards program is developing CdZnTe detector technology in collaboration with industry, other DOE facilities, and the IAEA. This includes research on device physics and electronic properties and the development of specialized detection probes, electronics, and spectrum analysis software. Implementation of the technology is facilitated by two OSS-funded projects: one to develop analytical methods and software for spectrum analysis; and the other to develop high-efficiency, hand-held spectrometers. User-friendly spectrum analysis software is being developed for field use with large-volume CdZnTe detectors. This software will fill a variety of functions for safeguards, including data acquisition from portable MCAs, isotope identification, peak-area determination, and determination and verification or isotopic composition and enrichment.

The detector-development project is addressing the need for low-power temperature control for measurements at elevated ambient temperatures, as well as new methods to improve detection efficiency and performance. In parallel, we are developing compact detection probes for the IAEA. This effort has resulted in a hand-held detector that compares favorably in terms of efficiency with the largest NaI(Tl) detector used for holdup measurements. We are also developing CdZnTe-based, gamma-ray spectroscopic imaging for holdup measurements and miniature, autonomous CdZnTe sensors for process monitoring, and compact Compton suppression systems to improve the sensitivity of hand-held CdZnTe detectors.

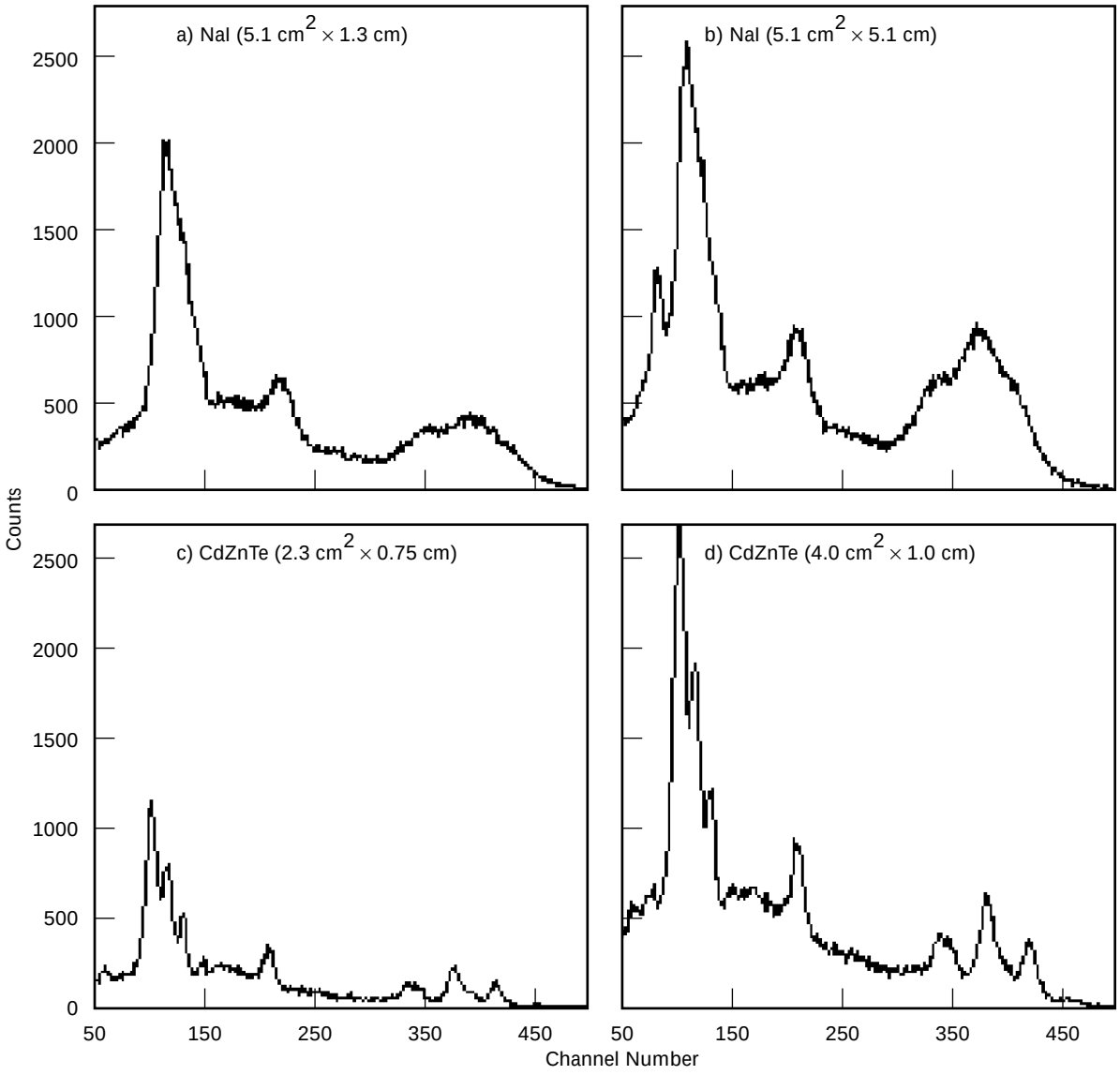

Figure 1. NaI(Tl) detectors, commonly used for holdup measurements and in situ NDA, are contrasted with CdZnTe detectors. The measurements were made under the same conditions: the source was 5 g of low burnup plutonium; all measurements were made with the same count time (100 seconds) with the source placed the same distance from the detector and with the same thickness low-energy filter. The spectrum in (a) was taken by the standard NaI(Tl) detector used for uranium holdup measurements; (b) by the standard NaI(Tl) detector used for plutonium holdup measurements; (c) w by the largest practical single-crystal CdZnTe detector; (d) by a large-volume, hand-held multiple-crystal CdZnTe detector developed by Los Alamos National Laboratory in collaboration with eV Products. The dimensions of the detectors (area × thickness) are given. Note that channel number is roughly proportional to gamma-ray energy in keV.

Electro-Mechanically Cooled Germanium Detector System

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The performance and reliability of germanium detectors, well known for analyzing special nuclear materials (SNM), have become the standards for all gamma radiation detectors. Combining high performance with high efficiency, germanium detectors provide very high-resolution radionuclide spectra in minimal time. Many software codes take advantage of germanium detector spectra (MGA, MGAUN, GRPANL, etc.), several of which have also become industry standards.

Germanium detectors have historically been relegated to the laboratory because of the need for extensive support equipment, typically including signal-processing electronics for data acquisition (shaping amplifier, test pulser), a high-voltage power supply for detector bias, a multi-channel analyzer (MCA), a computer including requisite software necessary for spectral analysis, and (most importantly) a continuous supply of liquid nitrogen to maintain the detector at its proper operating temperature. Clearly, the logistics of field applications are problematic.

Figure 1. The portable Electro-Mechanically Cooled Germanium (EMC-Ge) detector system uses a Stirling refrigerator.

As the need for field detectors increased, many support elements are now available as portable systems. Integrated electronic systems are currently available that, along with a detector and portable computer, provide portable data acquisition. Examples include the MiniMCA (GBS Elektronik), the M^3CA (Los Alamos National Laboratory), the InSpector (Canberra), and the Dart, MicroNOMAD, and NOMAD (Perkin-Elmer ORTEC). In addition to the reduced size, weight, and power requirements of the detector electronics, several manufacturers have developed small liquid-nitrogen dewars. These portable dewars typically hold enough liquid nitrogen for 12 to over 36 hours. (The typical germanium detector consumes approximately 1.5 liters/day.)

Although portable dewars help immensely, many applications require long acquisition periods, unattended operation, or an elevated temperature environment that rapidly depletes the gas supply. Clearly, a refrigeration system is needed that provides the cooling and operates reliably and independently, with a minimum amount of maintenance and power consumption.

The battery-operated Electro-Mechanically Cooled Germanium (EMC-Ge) Detector System uses a mechanical cryocooler to cool the germanium crystal (**Figure 1**). Active

vibration dampening reduces the deleterious effects of mechanical vibration. The detector system is unique because it is portable, requires little or no maintenance, consumes low amounts of power, and has a long operational life.

The EMC-Ge system contains a linear-motion Stirling refrigerator that uses a patented gas-bearing technology to virtually eliminate mechanical friction, greatly decreasing wear and increasing lifetime. A motor-driven counterbalance is mounted on the same axis as the refrigerator to attenuate vibrations that normally degrade the signal from the detector. A digital signal processor drives the counterbalance based on internal accelerometer readings and the system's vibration characteristics. Requiring less than 100 W of 12-Volt DC power, the EMC-Ge can be operated from a battery, a car cigarette lighter, or via an AC-to-DC adapter.

Several systems have been tested that included various detector volumes ranging from small planar detectors (Ortec LEPS and Canberra LEGE) to large 55% relative efficiency coax Ge detectors. Each system presented new challenges to realize optimum performance relative to energy resolution, power consumption, and overall system stability. The system that employed the 55% coax detector has been deployed in numerous demanding applications: high temperature and high external vibration and shock environments, and applications requiring remote and autonomous operation. In each deployment, the detector system performed without failure.

This detector system, combined with a custom software analysis package, was successfully demonstrated to U.S. Customs as a rapid isotopic identification system. Named the Field Radionuclide Identification System (FRIS), the system identified all the unknown radioactive sources in the general vicinity of the detector within 3 minutes. This is a significant achievement, in that all previous systems (most of which are sodium iodide-based) could not accurately provide this information unless the source was monoenergetic or completely characterized and known. Two planar detector systems were delivered to the European Communities' Safeguards Directorate (EURATOM) and the detector development group at the International Atomic Energy Agency (IAEA) for use in unannounced inspections and unattended or portable monitoring activities (**Figure 2**).

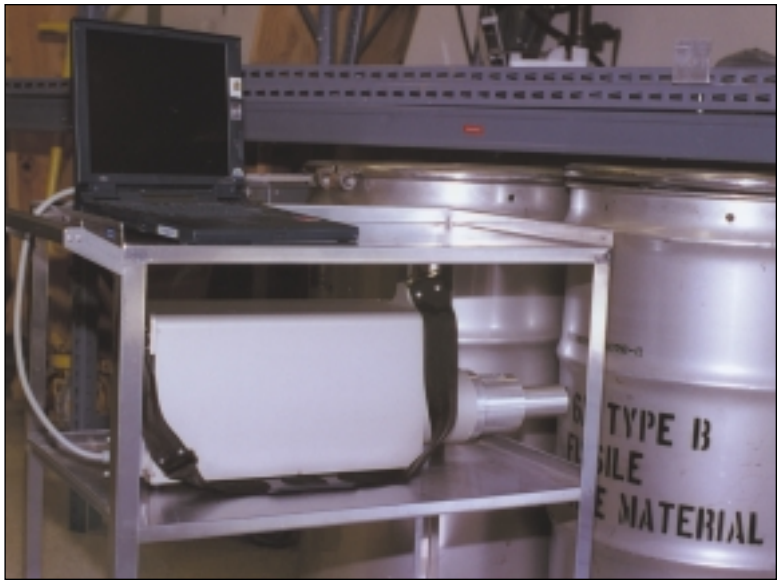

Figure 2. The EMC-Ge detector system can be left unattended.

Spectroscopic Imaging for On-line Measurements of Plutonium Inventory

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A new, commercial gamma-ray spectroscopic imaging system with a coded aperture is being tested on line at the Los Alamos National Laboratory's TA-55 facility. The inventory of plutonium in a cascade dissolver will be evaluated dynamically as it operates. Materials include plutonium, americium, and other elements in changing amounts and distributions. The plutonium inventory is normally determined by a time-consuming cleanout of the equipment. By eliminating this step, productivity will be improved and costs reduced.

The imaging system operates interchangeably with either a pinhole or coded aperture. We are currently using a CsI(Na) scintillator with a position-sensitive photomultiplier tube. It produces a two-dimensional image of gamma rays emitted in the detector's field of view, displaying the gamma-ray image superimposed on the aligned photographic image of the equipment. The system "images" gamma rays in each of several selected energy regions. The first in-situ tests used gamma rays of ^{237}U and ^{239}Pu at 208 and 414 keV plus a reference peak (to establish gain and absolute response and for image alignment) at 662 keV. The 662-keV peak is also a high-energy signature of the dominant isotope ^{241}Am.

The three cascade dissolver columns, along with equipment related to the dissolver system, operate within a glove box (**Figure 1**). Oxide and acid are fed continuously into the first column, which overflows into the second, and the second into the third. Gamma

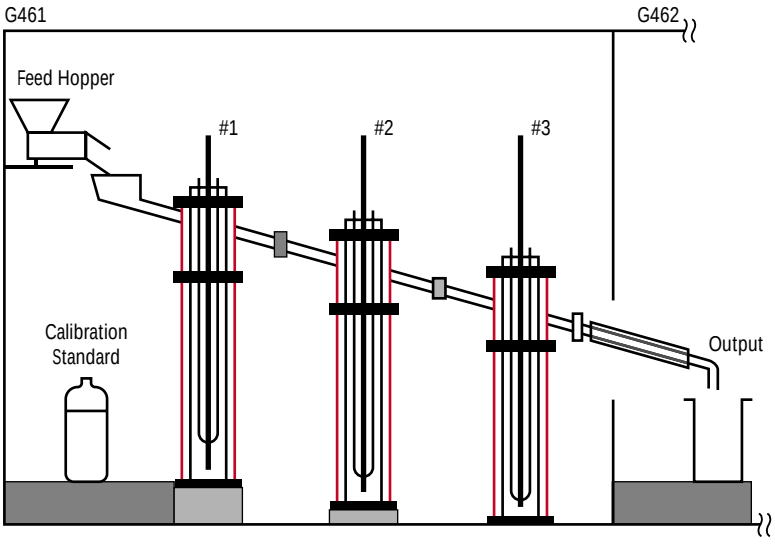

Figure 1. The three-cascade dissolver columns, outlined in red, are located inside the glove box. The bottle in the lower left corner contains a reference solution of plutonium whose spectroscopic image and known concentration provide an in-situ calibration of the gamma-ray measurement.

Figure 2. The spectroscopic imaging system is installed and operating in the process area of TA-55. The measurement head of the imaging system is at the center of the photograph mounted on a plate that rests on the distillate storage tanks. The aperture is aimed at the dissolver equipment within the glove box directly across the aisle (left side). The data processing equipment and automation software reside within the PC. No other equipment is required.

rays emitted by material within the dissolver are detected if they penetrate the glove box's walls. Higher energy (more penetrating) gamma rays are therefore selected.

Figure 2 shows of the gamma-ray spectroscopic imaging system on line at TA-55 opposite the cascade dissolver's glove box. The system's plutonium loading and distribution were varied during the measurements to evaluate the measurement sensitivity and spatial resolution on line.

Figure 3 shows a coded-aperture image of the ^{239}Pu gamma ray superimposed on the photograph of the dissolver in its glove box. The red outlines mark the columns' positions within the glove box, indicating that ^{239}Pu is concentrated in the second and third columns. However, this spectroscopic image still requires two corrections: one for the geometric response to a distributed source and another for gamma-ray attenuation by the glove box materials.

The two-dimensional, geometric-response matrix has been determined experimentally. For attenuation, corrections have been calculated. Matrix positions coincide with the pixels in the 38-by-38 spectroscopic images. Corrected spectroscopic images of the reference solution and dissolvers will be used to quantify plutonium in the dissolver. The spectroscopic imaging system will soon be upgraded to acquire three parameter (x and y positions and gamma-ray energy) data for more complex spectral analysis and simplified on-line measurements.

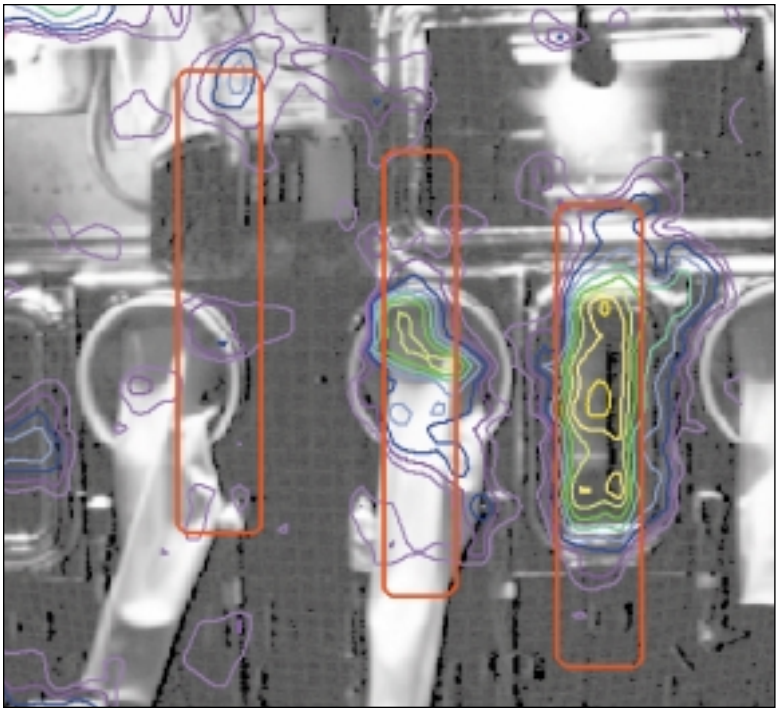

Figure 3. The coded-aperture spectroscopic image of the 414-keV gamma ray from ^{239}Pu is superimposed on the photographic image of the glove box. The positions of the three-cascade dissolver columns inside are outlined in red, analogous to the outlines in Figure 1. The spectroscopic image data have not been corrected for geometric response and gamma-ray attenuation.

Advanced Isotopic Analysis Software

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The DOE laboratories develop gamma-ray analysis codes to determine the isotopic abundances of special nuclear material. MGA++, developed by Lawrence Livermore National Laboratory, is a suite of computer codes that analyzes gamma-ray spectra and determines the relative isotopic abundances of actinides in a sample. The code suite analyzes data collected with high-purity germanium systems for plutonium and uranium isotopes (**Figure 1**). Additionally, the code suite can analyze uranium spectra collected with cadmium-zinc-telluride (CdZnTe) detectors. PC/FRAM, developed by Los Alamos National Laboratory, determines the isotopic composition for plutonium and uranium using data collected with a high-purity germanium detector.

The original MGA code was developed to determine plutonium isotopic abundances for gamma-ray data collected with germanium detectors. The MGA++ code suite includes MGA as well as several other codes for isotopic analysis of uranium and plutonium samples. The MGA++ code suite is fully modularized. All codes in the MGA++ code suite are self-calibrating. Typical count times are 5–30 minutes, and the analysis requires a few seconds of computer time. PC/FRAM requires calibration by the user. Typical PC/FRAM counts times are several hours.

The first code in MGA++ is an upgraded version of MGA that analyzes the plutonium isotopics. This upgraded version has been supplied to Euratom in Luxembourg for European safeguards work (**Figure 2**). The MGA code is currently used at many sites in the U.S., Europe, and Russia. MGA accurately analyzes plutonium–uranium mixtures. U235 is a uranium isotopic analysis code that uses gamma rays and x rays to determine uranium isotopic abundances (**Figure 3**). CZTU is a uranium isotopic analysis code that uses gamma rays collected with

Figure 1. Two high-purity germanium detectors collect gamma rays from a plutonium sample. Data are analyzed to provide plutonium isotopic information.

a room temperature CdZnTe detector. Pu600 is a plutonium isotopic analysis code that uses the 600-keV to 700-keV energy region.

Recently, a new code has been added to MGA++. MGAHI is a plutonium isotopic analysis code based on MGA. The MGAHI analysis is similar to MGA but it has been improved to overcome the limitations encountered by MGA with shielded samples and samples in a hot, space-limited environment. Future development of MGA++ will include isotopic analyses on mixtures of plutonium and uranium. We are also developing a new code that will analyze plutonium isotopics from data collected with CdZnTe detectors.

Several of the codes have been developed via a cooperative R&D Agreement (CRADA) between Lawrence Livermore and ORTEC: MGA, U235, and CZTU. Also, CZTU has also been licensed to Canberra Industries. PC/FRAM has been licensed to ORTEC and to Canberra Industries.

While MGA++ and PC/FRAM are limited to analyzing actinides, other codes have been developed that analyze any radioactive sample, including samples about which nothing is known. These general-purpose codes, including GAMANAL, have very diverse applications including Comprehensive Test Ban Treaty scenarios, nuclear forensics, environmental monitoring, and airport security.

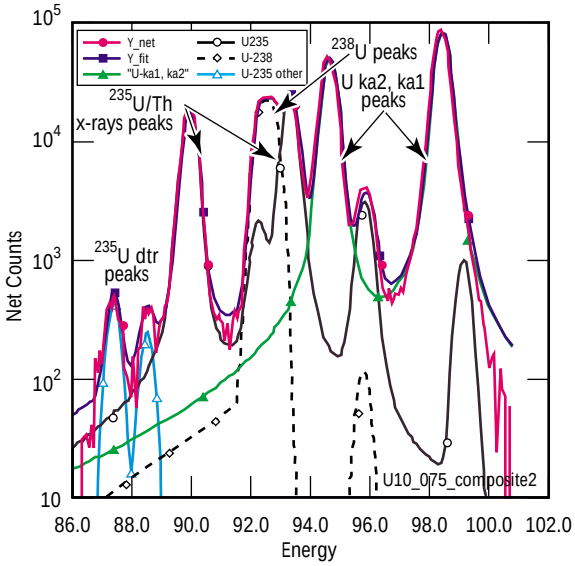

Figure 2. Data collected with a germanium detector has much sharper peaks, so the U235 code produces more accurate uranium isotopic information.

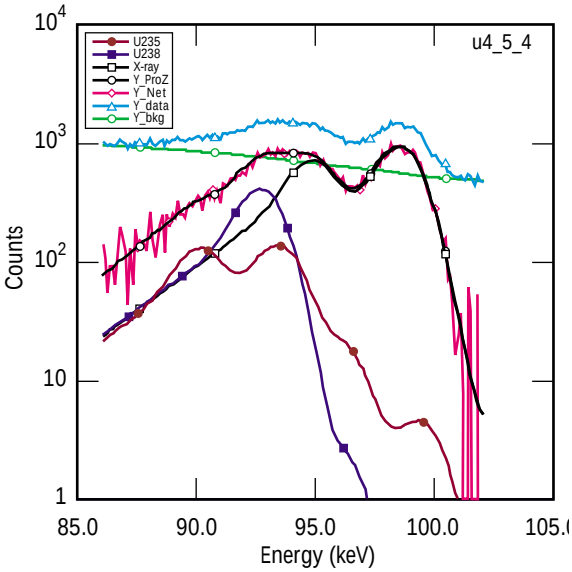

Figure 3. This uranium isotopic analysis shows the broad peaks characteristic of a room-temperature cadmium-zinc-telluride detector.

The FRAM Software for the Nondestructive Isotopic Analysis of Actinides

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Los Alamos National Laboratory's legacy as the developer of first gamma-ray analysis methods for measuring the isotopic composition of plutonium in arbitrary samples continues with its FRAM software. FRAM uses a single detector—either a planar or coaxial detector—to acquire data, making FRAM systems inherently more versatile, more reliable, easier to use, and less expensive.

Early on, materials were measured exclusively with planar detectors. As requirements changed, Los Alamos developed the methods that relied mainly on more efficient coaxial detectors. Using FRAM with coaxial detectors has opened up the use of gamma-ray isotopic analysis software for areas unheard of just a few years ago:

- uranium isotopic analysis of arbitrary samples
- complete plutonium isotopic analysis through lead or highly attenuating containers
- complete uranium isotopic analysis through thick UF_6 cylinder walls;
- "on-the fly" modifications to account for unexpected measurement conditions
- analysis of shielded or unshielded plutonium, shielded or unshielded uranium with a single detector, a single program, and a single data-acquisition setup.

FRAM is extremely user-friendly. All parameters governing data acquisition and analysis are placed into a database that users can edit. These parameters may be changed from within a password-protected option with the

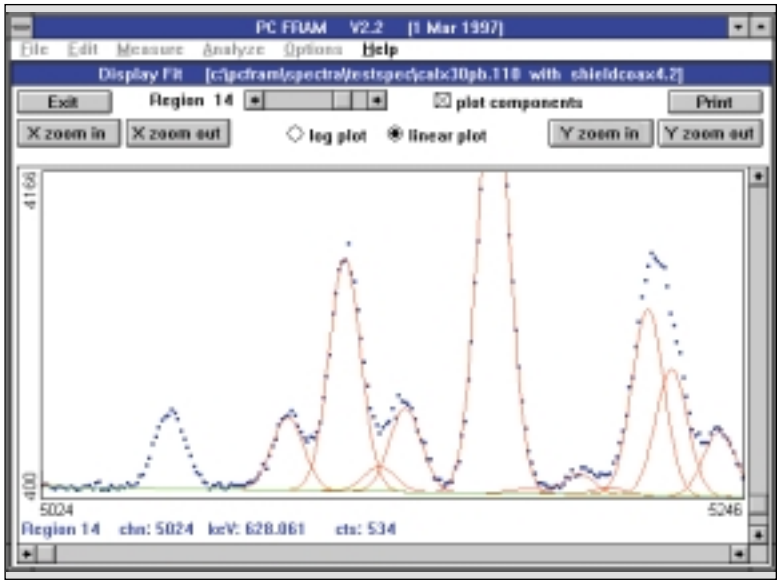

Figure 1. The FRAM software allows users to view detailed results of the peak fitting in all of the analyzed spectral regions. The fitting in the 640-keV region as displayed by FRAM is shown from the analysis of a lead-shielded sample of weapons-grade plutonium.

look and feel of a spreadsheet. Standard sets of parameters accomplish all routine assays. Versatile display features allow users to view important intermediate results such as peak fitting (**Figure 1**) and the relative efficiency curve (**Figure 2**).

FRAM's flexibility allows the analysis of the widest range of material categories possible. FRAM, with a single coaxial detector and a single set of data-acquisition conditions (no electronics changes between material categories), can measure all of the categories shown in **Table 1**.

The FRAM isotopic analysis software, available commercially from three licensees, is certified for use by the IAEA. It is also certified for the Waste Isolation Pilot Plant. FRAM is used in Russia and is planned for use in the Russian weapons plutonium conversion line to mirror its use in the ARIES weapons component dismantlement line at Los Alamos. The ARIES line recently completed an extensive demonstration run in which FRAM measurements coupled with calorimetry measurements of the product plutonium oxide showed agreement with accepted values for total plutonium mass to 0.05%.

In other projects, we are characterizing the performance of FRAM in measuring uranium in thick-walled UF_6 cylinders using planar detectors, often thought to be too inefficient at high energies for these measurements. We are also studying measurements on certain types of weapons components. FRAM is available with built-in motor control to position sample detector and interface the sample platform with robotics.

Table 1. Material Categories Analyzed with FRAM.

- **Plutonium with 0.03–99.9% ^{240}Pu**
- **Plutonium with nonequilibrium ^{241}Pu-^{237}U**
- **Uranium (only) ^{234}U, ^{235}U, ^{238}U with 0.2% < ^{235}U < 99%**
- **80% ^{238}Pu (heat source grade)**
- **MOX (mixed U-Pu oxide) with ^{235}U/Pu from 0.005–35**
- **Lead-shielded samples, up to 1 inch (25 mm) for plutonium**
- **Interferences from ^{243}Am–^{239}Np, ^{237}Np, ^{244}Cm, common fission products**
- **Complete uranium isotopic measurements through up to 17-mm steel (> 0.2% ^{235}U) and 1.6 mm Pb (> 20% ^{235}U)**
- **0.01–80% ^{241}Am**
- **Heterogeneous Am/Pu (pyrochemical residues)**

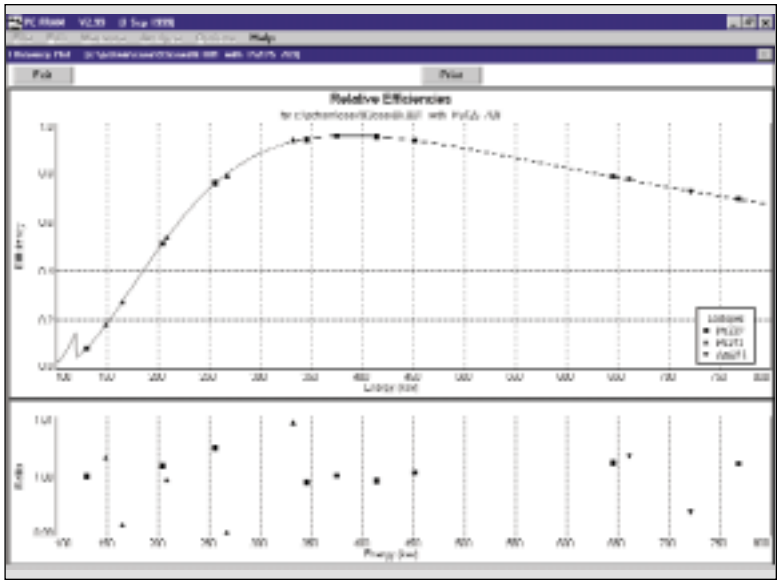

Figure 2. The fitting to the relative efficiency curve as displayed by FRAM.

CZTPU Software

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CZTPU is a computer code that, upon completion of its development, will analyze gamma-ray and x-ray data collected nondestructively with a cadmium zinc telluride (CdZnTe) detector, providing the user with information about the isotopic composition of plutonium in a sample.

CdZnTe is a crystal that detects gamma rays and x rays emitted from radioactive materials. CdZnTe detectors have a resolution midway between that obtained with sodium iodide and germanium crystal detectors. CdZnTe detectors operate at room temperature; they may be cooled by mechanical means, generally Peltier cooling. A Peltier-cooled detector offers better energy resolution than a room-temperature CdZnTe detector, so subsequent spectral analysis is more accurate (**Figure 1**).

The CdZnTe detector resolution is significantly improved over that of sodium iodide detectors, allowing much more detailed isotopic analysis of uranium samples. The most serious limitation of CdZnTe detectors is their extremely small detector volume that limits their ability to obtain good counting statistics in a reasonable amount of time from most samples. Because of their small size, however, CdZnTe detectors are extremely portable.

In spite of this limitation, significant progress has been made in developing software to analyze the data collected with CdZnTe detectors. The CZTPU code analyzes data between 50 and 200 keV and determines the relative isotopic ratios of ^{235}U, ^{238}U, and (to a smaller level of accuracy) ^{234}U in uranium samples. Also, CdZnTe identification software distinguishes between uranium, plutonium, and neptunium.

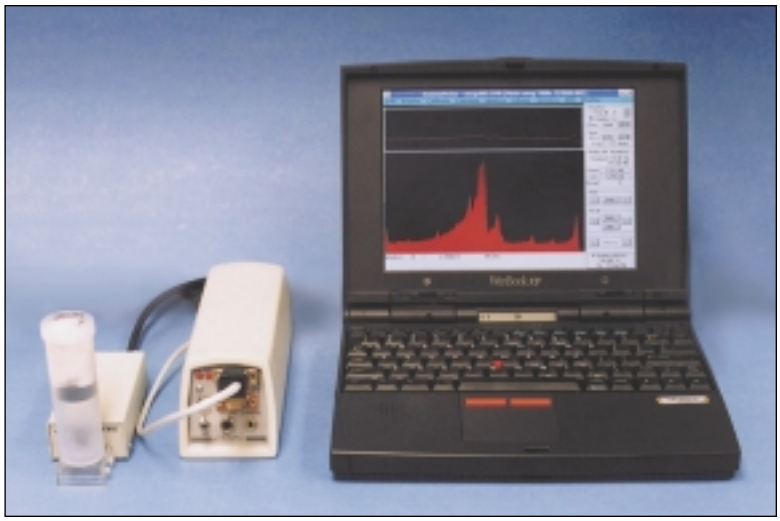

Figure 1. A laptop runs the CZTPU software. The detector is on the tip of a preamplifier (behind the sample in the plastic tube). The system also includes a multichannel analyzer.

The plutonium isotopic determination must rely on the 50- to 200-keV energy range because this is the limitation of CdZnTe detectors currently available. The most useful plutonium peaks lie in this energy range, but the peaks overlap and the energy region is extremely complex. The possibility of developing a code that uses data collected with room-temperature and Peltier-cooled detectors is being investigated. The final version of the software will distinguish between weapons-grade and nonweapons-grade plutonium. It is also possible that the final code will state the relative isotopic abundances of ^{239}Pu, ^{240}Pu and ^{241}Pu—within a predefined level of accuracy such as 20%. This second solution may rely on the use of the Peltier-cooled system.

Development will proceed in two phases. Phase I includes acquisition and characterization of a typical CdZnTe detector system as well as acquisition of a basis set of gamma-ray spectra of known plutonium sources as measured with a well-characterized CdZnTe detector. The alpha release code will be based on these measurements. Phase II involves strengthening the CZTPU alpha release code by incorporating spectra of known plutonium sources as measured with other CdZnTe detectors. Phase II will also involve necessary refinements to optimize the final solutions, and pursuit of licensing agreements with commercial companies to make the software easily available.

NDA of Uranium in Weapons Components and Mixed-Oxide Fuels

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The disposition of excess weapons components from the U.S. stockpile includes the conversion of plutonium and uranium metal into oxides for mixed-oxide (MOX) fuel for commercial nuclear power reactors. There is already experience with NDA measurements on plutonium from weapons components, but no experience with the uranium components. Uranium needs an active neutron technique. In this study, all the options will be explored and a recommendation will be made. The variety of geometric forms needs to be studied to incorporate variations in calibration with shape as well as with the mass. Also, the presence of other materials in a weapons component complicates active-neutron measurements. Our study combines measurements on weapons components and the calculations for shape (**Figures 1** and **2**).

Eventually, weapons materials will be formed into MOX fuel. MOX is already used in other countries, but the U.S. has little experience with it. MOX fuel from the U.S. will be quite different isotopically from the rest of the world's because it comes from weapons rather than from reactors. The experiences of other countries in safeguarding MOX are useful, but the impact of different isotopes has to be taken into account.

This project will help close the material balance around the disposition process. The first step is a clear understanding of how to measure uranium metal in a variety of conditions previously unmeasured. Next, the impact of weapon-grade isotopes on the measurements of MOX will be studied. The interactions of different fissile materials, non-fissile materials, and impurities will also be clarified.

Key Milestones	Completion Date
Complete initial investigation of candidate technologies for measuring uranium metal from components	1/1/99
Complete an assessment of component-type calibration dependence of candidate techniques	3/1/99
Complete initial Monte Carlo calculations to understand the coupling between interrogating source neutrons and induced fissions	5/10/99
Report on the results of investigations and calculations to date	9/1/99
Report on candidate detector technologies that will be successful in the measurements of uranium metal from components	2/1/00
Complete a study of candidate technologies for the measurement of MOX materials	6/1/00
Report on potentially successful measurement techniques for MOX mixtures	8/1/00
Complete a study of the effects of different MOX compositions on the multiplicity assay of impure MOX scrap and begin to work toward mitigating these unwanted effects	1/1/01
Report on the study of effects of different MOX compositions and how to mitigate their effects	8/1/01
Complete proposals of candidate instrument technologies that will provide the best NDA measurements for MOX mixtures and fuel configurations	1/1/02
Report on the conclusions from the project	8/1/02

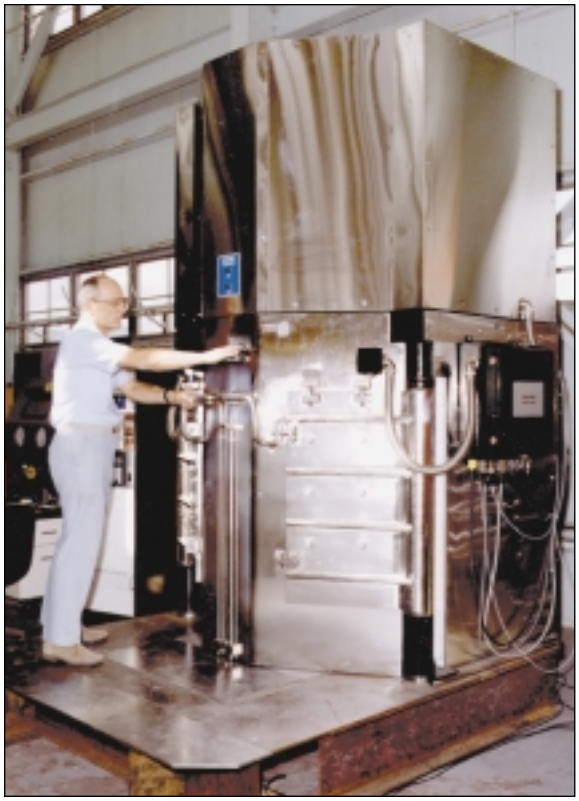

Figure 1. A strong candidate for assaying the uranium in weapons components and MOX fuels is the shuffler. The general-purpose shuffler shown here is large enough to hold a 55-gallon drum. This size and the strength of the ^{252}Cf source can be shrunk to better match the sizes of components and MOX containers. The assay chamber is the lower half of the instrument and the ^{252}Cf source is stored in the shield that makes up the upper half.

Figure 2. This passive multiplicity counter was built by Los Alamos National Laboratory for 30-gallon drums stored at the Rocky Flats facility. It could measure the plutonium in weapons components and in MOX containers. Smaller counters are routinely used for can-sized containers.

Verifying the Enrichment of Gaseous UF_6 with the Cascade Header Enrichment Monitor

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The International Atomic Energy Agency (IAEA) inspects gas-centrifuge facilities in Japan, England, Germany, and the Netherlands. The objective of these inspections is to verify the absence of highly enriched uranium, and that the product material in the enrichment facility is consistent with the operating license of that facility. These inspections are called Limited-Frequency Unannounced Access (LFUA). A limited number of inspections can occur during the year, and the host facility is notified a relatively short time before the IAEA inspectors appear. The Cascade Header Enrichment Monitor (CHEM), developed by the Los Alamos National Laboratory, is approved for an LFUA by the IAEA. The CHEM was also used by the IAEA in 1998 during their verification experiment at the Portsmouth Gaseous-Diffusion Plant in Piketon, Ohio, to confirm that the U.S. did blend down highly enriched uranium to low-enriched uranium.

To determine the enrichment of the gaseous UF_6, CHEM needs two quantities: (1) a measure of the amount of ^{235}U present in the gas, and (2) a measure of the amount of total uranium present in the gas. CHEM determines the amount of ^{235}U from the intensity of the 185.7-keV gamma ray from the decay of ^{235}U. The total amount of uranium is determined by using a ^{57}Co x-ray fluorescence (XRF) source to excite the 98.4-keV uranium x ray. The ratio of these two quantities is directly proportional to the enrichment of the uranium in gaseous UF_6.

The formation of uranium deposits on pipe walls complicates this procedure. The intersection of the solid angles of the detector collimator and the XRF source collimator and the relative angle between the detector and

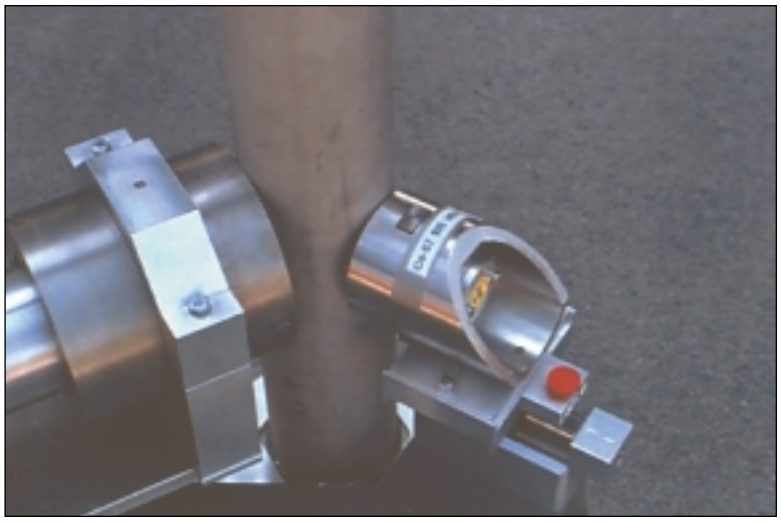

Figure 1. The CHEM developed for the Rokkasho Enrichment Plant, Japan. The relative angle in the collimation of the XRF source and the gamma-ray detector eliminate the contribution from fluorescing the deposits of uranium.

the XRF source eliminate the contribution from fluorescing the deposit. **Figure 1** shows the relative angle between the detector and the XRF source for the CHEM developed for the Rokkasho Enrichment Plant in Japan.

However, the deposit directly in front of the detector contributes to the 185.7-keV gamma-ray intensity. One way to account for the deposit is to measure the background intensity directly while no gas is present in the pipe, and subtract it from the total 185.7-keV intensity. This method is not always possible. A second method corrects for the deposit without the need to measure the evacuated pipe. This second technique uses two collimators for two separate, independent measurements of the 185.7-keV gamma-ray intensity to measure the ^{235}U in the gas independent of the ^{235}U in the deposit. The collimator for the two independent measurements of the 185.7-keV gamma-ray intensity, again for the CHEM for the Rokkasho Enrichment Plant, is shown in **Figure 2**. The collimator is rotated through 90 degrees to effectively and efficiently generate the two measurement geometries. For a pipe that has very small deposits, a ratio of a single measurement of the 185.7 keV gamma-ray intensity to the XRF signal is sufficient for the enrichment measurement. The CHEM for the Rokkasho Enrichment Plant was installed and demonstrated during the summer of 2000.

The CHEM instrument is compact. Typical header pipe diameters are 8 cm to 11 cm. The CHEM uses a portable, high-resolution germanium detector, a laptop computer, and small pulse-processing electronics. It is easily operated by one IAEA inspector. The computer instructs the IAEA inspector on the correct steps of the measurement, collects and analyzes the data, makes all the decisions based on the data analysis, and informs the IAEA inspector of its conclusion.

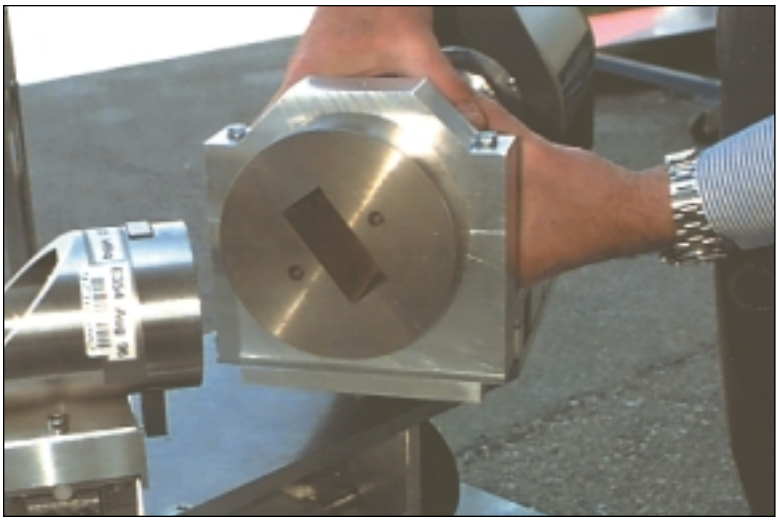

Figure 2. The collimator for the two independent measurements of the 185.7-keV gamma-ray intensity is shown for the CHEM for the Rokkasho Enrichment Plant, Japan. The collimator is rotated through 90 degrees to effectively and efficiently generate the two measurement geometries.

NDA Methods for Neptunium

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Many techniques have been developed in the U.S. and other countries to control the special nuclear materials (SNM), plutonium and uranium, but no standard methods exist to determine neptunium in bulk containers. Such methods are needed because the Department of Energy requires that all government-owned ^{237}Np be treated as if it were SNM. In particular, shipping and receiving and inventories need to be verified. We are developing passive and active techniques for neptunium metal and oxide.

The passive analysis of gamma-ray lines can be used if the material is not so heavily shielded preventing gamma-ray emission. Some shielding is often used for neptunium because the strong gamma rays from the daughter isotope ^{233}Pa can produce a significant radiation hazard. If gamma rays are emitted, analysis of passive gamma-ray spectra acquired with a high-resolution germanium detector (**Figure 1**) uniquely identifies the material, provides isotopic ratios for impurities such as ^{243}Am, and provides some information about the shielding, mass, and time since processing. Software is being developed to provide this information for neptunium.

A tomographic gamma scanner (TGS) can map the distribution and approximate dimensions of the neptunium and shielding in a container, again if gamma rays are emitted. A germanium detector measures the emissions from the daughter ^{233}Pa and transmission from an external source of ^{137}Cs. **Figure 2** shows four small samples of neptunium metal in a 55-gallon drum.

Figure 1. A portable germanium detector can measure gamma-ray spectra in the field, and thus avoid the complications of transporting neptunium.

Active-neutron interrogation techniques can be used even if no gamma rays are emitted. One such technique is the differential die-away technique implemented in a package monitor. The technique uses neutrons from a pulsed neutron generator and arrays of ^{3}He-filled neutron proportional counters to detect neutrons produced by fission in the neptunium. By measuring changes in the neutron time-correlated flux, the presence of both prompt and delayed fission neutrons can be easily detected. The response depends on the mass of neptunium as well as on the shielding.

Another active technique involves a linac. Pulses of 10-MeV photons induce fission in the neptunium and an array of ^{3}He detectors detects neutrons between the pulses. The data are analyzed to determine how many events produced one neutron and how many produced two neutrons. Only SNM produces two neutrons per event. As for the package monitor, the response depends on the mass of neptunium as well as the shielding.

For standard packages, one of these techniques can be calibrated to provide a mass. However, for non-standard packages, data from more than one of these techniques can be combined to obtain a mass estimate, depending on the specific application. All techniques provide some information about the material, but analysis by passive gamma-ray techniques is the most definitive.

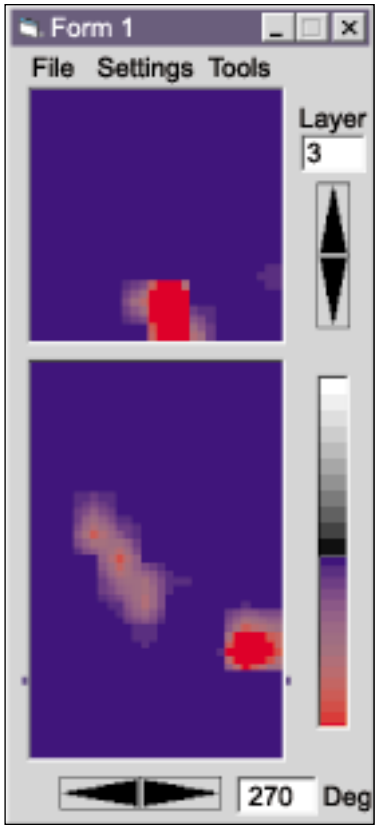

Figure 2. TGS emission image of four neptunium sources at different heights and radial positions within a 55-gallon drum. The upper image is a tomographic slice view; the lower image is a "radiograph" side view. One of the sources (the lowest in the drum) is approximately four times stronger than the other three and therefore appears larger on this intensity scale.

NDA of Impure Plutonium Metals and Oxides Stored in Shielded Containers

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Current plans for removing high-concentration plutonium metals and oxides from the Rocky Flats Environmental Technology Site call for these materials to be placed in 9,975 shipping containers, moved to the Savannah River Site, and then stored in their shipping containers at K-Area until final disposition. Shipment is scheduled to begin in FY 2000. Much of this plutonium is impure. The Rocky Flats shipper values will be based on calorimetry and isotopics measurements.

At Savannah River, the materials will remain in their drums for interim storage unless an anomalous condition is detected. The Westinghouse Savannah River Site has procured an integrated neutron multiplicity–isotopic measurement system for receipt measurements on these materials. Neutron multiplicity assay is the only option for these drums because of their size and insulating properties. However, recent experience with these materials has demonstrated that some technical challenges need to be resolved to ensure a successful outcome of this large material transfer.

The biggest challenge is measuring plutonium in a container specifically designed to reduce the very radiation emissions needed for a nondestructive assay. This has never been done before on the scale called for in the Rocky Flats plan. Experience measuring plutonium in shielded containers is very limited. Some measurements of plutonium in shielded AL-R8 containers illustrate the problem. **Figure 1** shows measurements of plutonium-

Figure 1. Measurements of plutonium-bearing materials in shielded drums at Rocky Flats Environmental Technology Site.

bearing materials in shielded drums being performed at Rocky Flats. **Figure 2** shows how assay results can vary for drums having diverse shielding characteristics. The ring ratio in this plot is a rough measure of the energy of the neutrons emitted and thus the effect of the shielding in a drum.

Another challenge is measuring impure metal samples. There is some experience measuring slightly impure metal with neutron multiplicity counting. This experience has resulted in the development of a correction algorithm to reduce assay bias. However, more recent experience suggests that if the metal is more than slightly impure, this algorithm is not adequate. **Figure 3** shows assays of slightly impure metals compared with metals having greater impurities.

This research effort, which began in FY 2000, focuses on calculations and measurements to understand the neutron and isotopic assay behavior of characteristic Rocky Flats materials in 9,975 shielded drums. We will work closely with both Rocky Flats and Savannah River personnel. Using data from the Savannah River equipment, we will develop algorithms to correct for drum-to-drum variability in the shielding materials to increase the accuracy of assays in shielded drums. We will also study the best analysis techniques for the neutron data to minimize biases, and methods that might be used to detect and resolve anomalies.

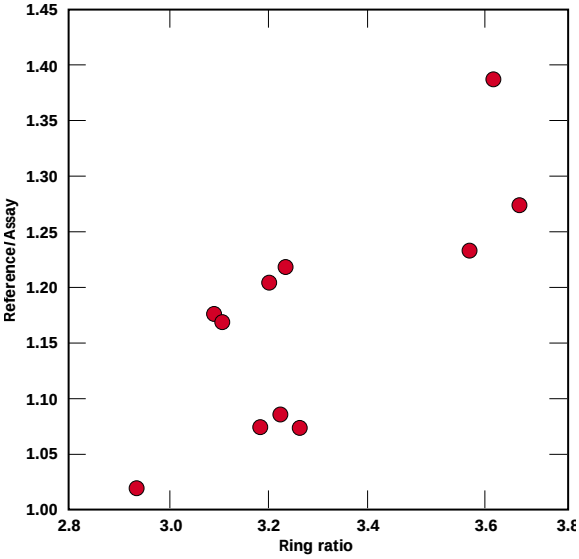

Figure 2. Drum-to-drum variability of assays of items in shielded drums.

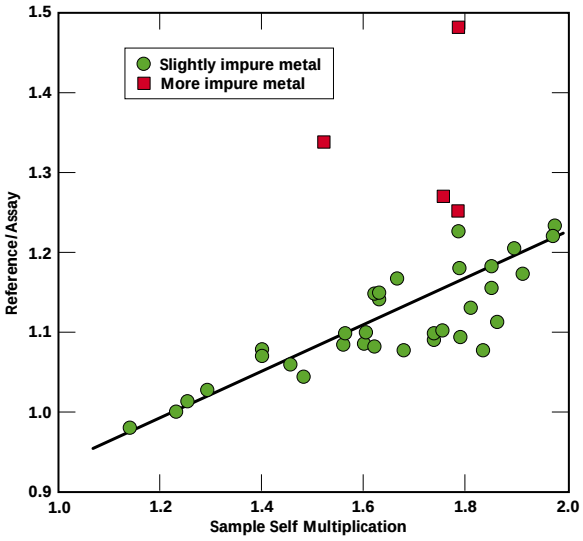

Figure 3. Neutron-multiplicity-assay results for slightly impure plutonium metal samples compared with samples containing large impurities. The current correction algorithm is not adequate for more impure metal samples.

Accountability Quality NDA Measurements on Pits

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The conceptual design for a Pit Disassembly and Conversion Facility (PDCF) included a technical risk assessment evaluating all of the processes in the facility for the risks associated with their inclusion in the facility. The ability of the facility to provide materials control and accounting (MC&A) measurements on the incoming pits was in the highest risk category because complete nondestructive measurements of all the special-nuclear-material masses in all pits scheduled for disassembly have not yet been demonstrated.

The pits in the U.S stockpile generally have either plutonium (weapons-grade) or plutonium and uranium. Our work concentrated on the latter case using an active/passive shuffler with multiplicity counting capability, and also on ways to improve the throughput for the former case with a gamma-ray isotopic measurement for ^{235}U-to-Pu ratio.

The shuffler measurements at Los Alamos' Plutonium Facility will combine active and passive techniques to unravel the signals from uranium and plutonium (**Figure 1**). Pits will be selected from those available through the surveillance program or from dismantlement. We will also investigate whether calorimetry measurements are required to assist in this process. Pits will be modeled using the MCNP Monte Carlo code to support and interpret the results.

Gamma-ray isotopic ratio measurements use a relative detection efficiency curve developed from the data for the pit being measured. Current methods do not work for all pits. We propose to develop an entirely new procedure for the relative detection efficiency, taking into account the known geometrical configurations of the pits.

We will incorporate these enhancements into a special version of the PC/FRAM isotopic analysis code and test it with the pits. This is the only code suitable for this work. Its modular structure and its current incorporation of the multiple relative efficiency curve technology will make this new approach easy to incorporate (**Figure 2**).

Milestones	Completion date
Begin measurements on pits	2/99
Begin computer modeling of pit neutron measurements	8/99
Complete shuffler measurements on pits	4/00
Complete measurements on pits	6/00
Complete report on pit measurements	7/00
Complete report on shuffler measurements of pits	7/00
Incorporate new relative detection efficiency into PC/FRAM	7/01

Figure. 1. A twin of this shuffler in Los Alamos' Plutonium Facility will be used for the neutron measurements on pits.

Figure 2. This type of gamma-ray system will be used for the isotopics study. The detector is adjusted easily to receive the best signal from the pit.

The NDA System for the ARIES Weapons-Component Dismantlement System

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ARIES (Advanced Recovery and Integrated Extraction System) is installed at the Los Alamos National Laboratory to demonstrate the technologies required to remove plutonium from surplus nuclear weapons and package it for long-term storage and ultimate disposition (**Figure 1**). ARIES is the basis for the designs under way for the Pit Disassembly and Conversion Facility (PDCF). The ARIES NDA system is also the design basis for the NDA system at the Russian Weapons Plutonium Conversion Facility.

The dismantlement process begins by introducing a weapon pit into ARIES. The pit is cut apart and the classified plutonium is converted to an unclassified form, as plutonium oxide or plutonium metal. This material is then packaged for long-term storage in a container meeting the requirements of DOE-STD-3013-96 (**Figure 2**). After the plutonium is packaged, the container is brought to the ARIES NDA system to measure and quantify the special nuclear material inside.

Figure 1. The ARIES nondestructive analysis system, installed at the Los Alamos National Laboratory's Plutonium Facility, has successfully completed an extensive demonstration. The system is now undergoing a series of planned upgrades before resuming operations.

Figure 2. Cutaway view of the DOE-STD-3013-96-compliant container used in ARIES showing the three levels of containment represented by the welded stainless-steel boundary container, welded stainless-steel material container, and the innermost convenience container.

The ARIES NDA system consists of three NDA instruments integrated with a central host computer and robotic sample handling. This automated system is capable of unattended operation round the clock, which increases the system throughput and greatly reduces material handling and thus radiation exposure to personnel.

The first NDA instrument is a calorimeter that measures the heat produced during the radioactive decay of the plutonium in the container. The amount of heat is related to the amount of plutonium being measured. Calorimetry is the single most accurate NDA method for bulk materials. The calorimeter in ARIES can measure the heat output from the plutonium in a container to an accuracy of better than one part in 1,000.

The second instrument, an active/passive neutron multiplicity counter, quantifies the plutonium by passively measuring the neutron output arising from the naturally occurring spontaneous fission. This same counter, used in the active mode, can also measure the mass of enriched uranium arising from the dismantlement process when the uranium is packaged in the same type of container. The robot automatically configures the counter for the proper measurement as shown in **Figure 3**.

The third instrument, a gamma-ray spectrometer, measures the isotopic fractions of the plutonium isotopes and americium in the sample. This information is used to convert the bulk measurement of heat (calorimeter) or neutron coincidence counts (active/passive multiplicity counter) into the plutonium mass.

All of the instruments and the robot are controlled by a central host computer. The host schedules NDA measurements, monitors the status of the instruments and containers in the system, commands the movements of the robot and commands the NDA instruments, as well as archives NDA measurements into its database. All of the instruments and host computer software were developed at Los Alamos and specially adapted to automated operation. The calorimeter uses the MultiCal control-and-analysis software. The FRAM isotopic analysis software is used for data acquisition and analysis giving plutonium isotopic fractions. The neutron coincidence counter uses the Los Alamos INCC code.

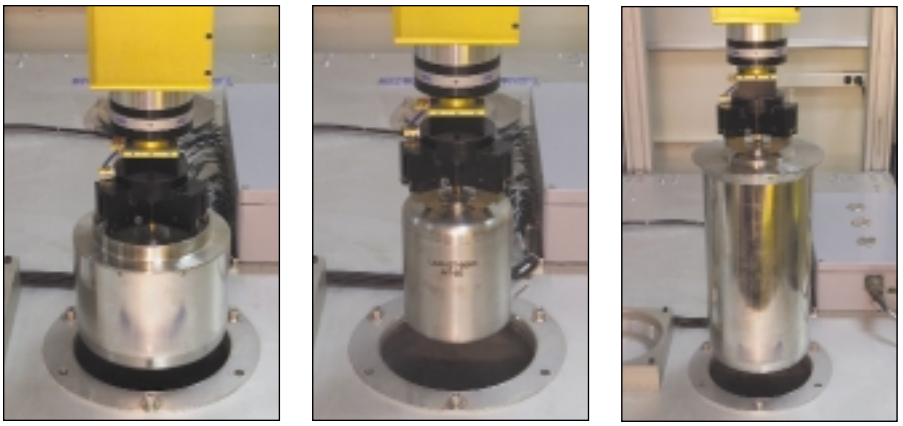

Figure 3. The robot loads the bottom end plug into coincidence counter (left), then loads sample container (middle), and finally loads top end plug into coincidence counter as last step before measurement (right).

Measuring the Plutonium Content of Spent Fuel at the BN-350 Reactor in Kazakhstan

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A large international safeguards program is presently under way to improve the safeguards and security of spent nuclear fuel from the BN-350 fast breeder reactor in Aktau, Republic of Kazakhstan (**Figure 1**). To satisfy International Atomic Energy Agency (IAEA) and Kazakhstan Atomic Energy Agency (KAEA) requirements, the plutonium content of these fuel assemblies and their heat output must be measured prior to their repackaging and relocation to difficult-to-access, long-term storage. We have nondestructively assayed the plutonium content of fast-breeder-reactor spent-fuel assemblies while still underwater in their facility storage pond. This is the first time that the IAEA has applied their partial-defect test criteria to spent fuel. The extremely accurate NDA of the plutonium content of BN-350 spent fuel was feasible because the reactor physics and operation are such that even at the highest fuel burnup levels, the buildup of the curium isotopes remains very low. The spontaneous fission neutrons generated by the spent fuel are dominated by ^{238}Pu and ^{240}Pu. A negligible amount of the spontaneous fission neutrons are generated by ^{242}Pu and the curium isotopes.

Figure 1. From left to right: Konstantin Serednyuk (MAEC/Kazakhstan), Igor Bolshinsky (Argonne National Laboratory), and Parrish Staples (Los Alamos National Laboratory) prepare to install the spent-fuel container into the spent-fuel pond at the BN-350 reactor in Aktau, Kazakhstan.

In our measurement technique, the singles and doubles neutron rates are observed in a ring of ^{3}He tubes embedded in polyethylene surrounding the fuel assembly while underwater in the facility storage pond. The singles and doubles neutron rates are converted into the plutonium content of a given assembly. The main steps in determining the relationship of the neutron count rate to plutonium content were the experimental determination of our neutron counter's response to various arrangements of fresh mixed oxide (MOX) fuel; Monte Carlo N-Particle (MCNP) modeling of these fresh MOX-fuel experiments; MCNP modeling of various BN-350 assembly types; and calculations of the relative amounts of various neutron-emitting isotopes in the BN-350 spent fuel. The facility used operator records and a computer code to declare the plutonium content of the BN-350 spent fuel assemblies. Using limited facility information, the combined measurement and facility declaration error is ~8%. A simplified one-point measurement procedure leads to a combined measurement and facility declaration error of ~13%. The heat output of each assembly was determined by a correlation of the measured gross gamma-ray output to calculated heat output for a set of calibration assemblies whose properties were well understood. The gross gamma-ray measurement was performed with a ionization chamber located within the spent-fuel container.

Automated Information and Data Management Tool To Evaluate SNM Inventory Differences

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We are developing an information and data management tool that will automatically and quantitatively evaluate differences encountered in inventories of special nuclear materials (SNM). The inventory difference (ID) for SNM is the defined as the book inventory (previous physical inventory + shipment – receipts) minus the physical inventory. Both inventory and transfer terms include measurement errors. If we know all the inventory and transfer terms and the measurement uncertainty associated with each measurement system, we can apply statistical rules involving the variance of a sum to estimate the measurement-error standard deviation of the ID, σ_{ID}.

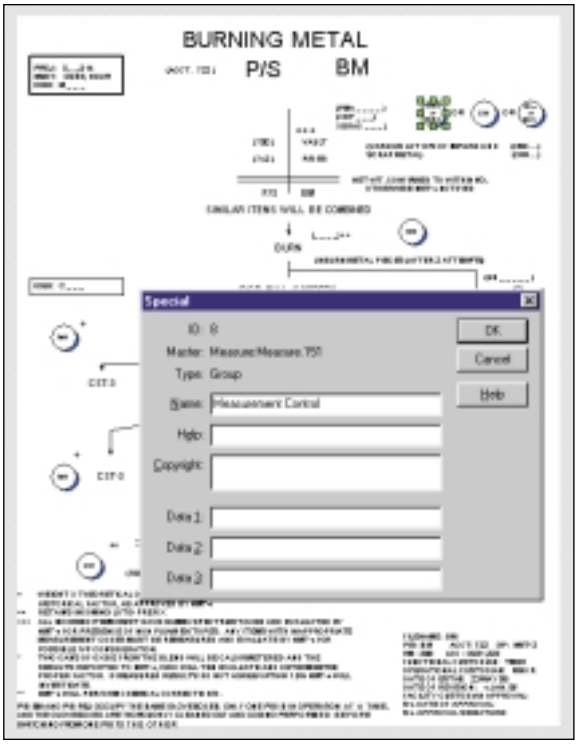

Figure 1. The flow diagram is shown for a particular process (Burning Metal). The green highlighted circle represents a link to other information/tools needed to evaluate inventory differences.

The ID calculation is the sum of many terms, some of which have negative signs. This project will support one way to quantify the effects (via propagation of variance) of systematic errors and random errors to estimate σ_{ID}. A good estimate of σ_{ID} is usually all that is needed to statistically evaluate an ID because of the central limit effect, whereby sums of approximately 10 or more random variables (all measurement errors are random at some stage, even the so-called "systematic" errors) will have approximately a normal distribution. Assuming no material loss, the ID has an approximate N(0, σ_{ID}) distribution [ID ~ N(0, σ_{ID})]. Therefore, to test for SNM loss, the ID can be compared to k σ_{ID} where k is 2 to 3, depending on the desired false-alarm probability.

Because we are using Los Alamos' Plutonium Facility for this tool, we chose a visual tool that will link to their visual process descriptions (process account flow diagrams). We selected the Visio enterprise commercial software to allow interaction with these flow diagrams (**Figure 1**). Visio is sufficiently versatile that we anticipate a relatively easy migration to other DOE facilities. The "interactive" flow diagrams allow the user to initiate any executable code. We plan to call Visual Basic® codes to interface to Microsoft Access® to estimate measurement-error standard deviations and perform variance propagation.

Milestones

FY 2000: Develop draft version of software tool, working with the Plutonium Facility at Los Alamos National Laboratory

FY 2001: Identify other potential DOE users and iterate with Los Alamos to modify the tool as necessary.

FY 2002: Final iteration with all users before they obtain control of the source code, and final report.

Local Area Network Materials Accounting System

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Since the inception of nuclear weapons, the U.S. policy—established by the Atomic Energy Act of 1954—has been to closely control the key materials for producing and developing nuclear weapons. With the advent of computers, the business processes used for this purpose were quickly computerized. Historically, inventory procedures were highly customized, with varied approaches, which made corporate management of the nuclear inventory difficult.

DOE sponsored the development of LANMAS, or Local Area Network Materials Accounting System, to standardize inventory and accounting across the complex. LANMAS tracks inventory at the item, container, or assembly level. It maintains historical audit trails for all changes to the inventory. Developed through teamwork and collaboration between DOE and its contractors, LANMAS has been successfully implemented at 9 DOE locations to date, with 15 sites expected by the end of FY 2001. Through standardization, LANMAS is laying the framework for today's business needs, which require more timely and complete nuclear-inventory information.

Originally, nuclear-material-accounting applications were derived from the paper-based business processes of the '50s and '60s. The data gathered was based on the information necessary to produce nuclear weapons. Today's management objectives focus on the disposition of these legacy materials. Environmental factors surrounding disposition require rethinking what information is needed. To meet this need, the underlying structure in LANMAS was engineered on traditional business, chemistry, and physics principles that support both the original nuclear-weapons objective and the new disposition requirement.

From LANMAS, methods to more efficiently manage inventory information on a corporate level have also emerged. In March 2000, the Westinghouse Savannah River Company, the current contractor responsible for LANMAS, and the Microsoft Corporation entered into a Cooperative Research and Development Agreement, or CRADA. The objective of the CRADA is to prototype state-of-the-art data management tools for the nuclear-inventory information within the LANMAS application from multiple locations. In four weeks, a prototype data warehouse merged the nuclear-material inventory data at the item level from four DOE locations (Argonne East, Oak Ridge National Laboratory, Mound, and Westinghouse Savannah River) into a single data mart for analysis and reporting.

LANMAS is a Client Server application written in Visual Basic® and Microsoft SQL Server® (**Figure 1**). It is scalable, supporting a corporate network or single personal computers. It accommodates 100 inventory items as easily as 1,000,000 items. It meets all current U.S. domestic reporting requirements (DOE 474.1-2).

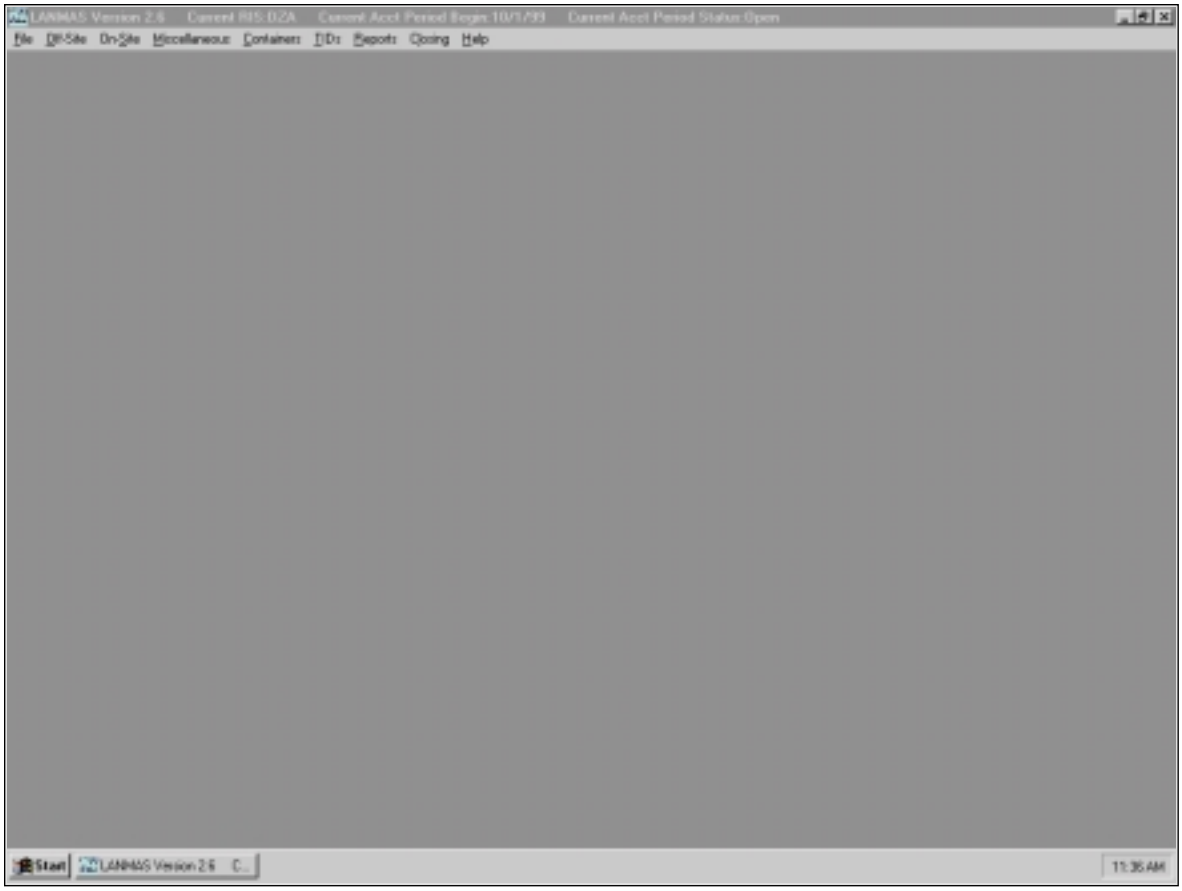

Figure 1. Sample screen from the LANMAS software.

Shielded Measurement System for Irradiated Nuclear Fuel Measurements

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Irradiated nuclear fuels (INF) are in long-term storage at various facilities throughout the Department of Energy complex. In the past, sampling and analytical chemistry verified the nuclear material inventory during reprocessing. In recent years, INF reprocessing has been discontinued, and thus the opportunity for sampling is more limited and therefore more costly. As a result, the nuclear-material inventory has been determined largely through calculations. In many cases, these calculations are not validated nor is the resultant inventory estimate validated through direct measurement.

In response, the Office of Safeguards and Security has promulgated requirements that INF inventories be determined by measurement or other validated means. Currently, Argonne National Laboratory–West is developing the Shielded Measurement System (SMS) to nondestructively measure the nuclear-material inventory at Argonne West. The system may prove to be applicable to INF stored at other DOE facilities.

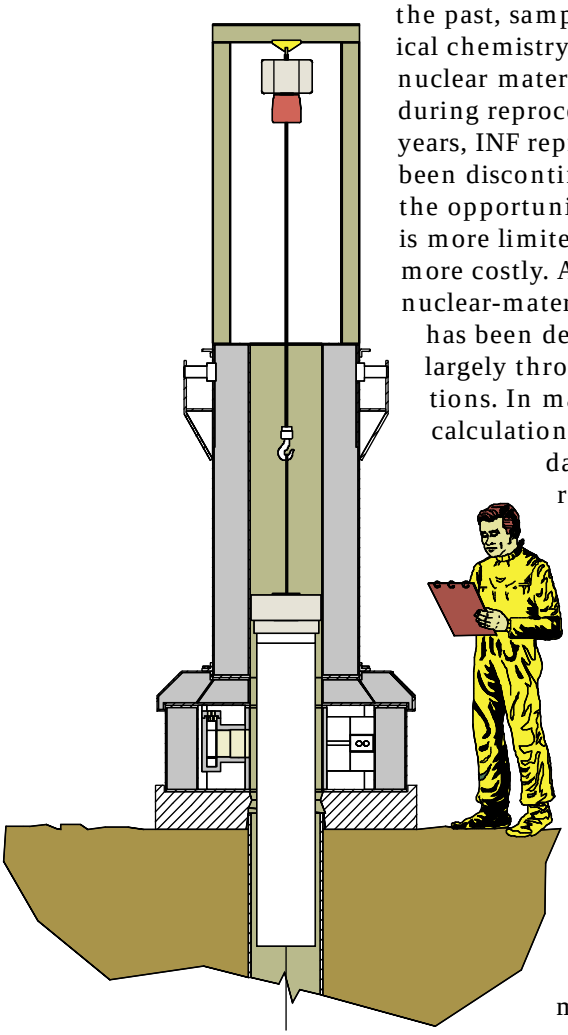

Figure 1. Sketch of the Shielded Measurement System shown measuring a waste can stored underground. The axial-scanning hoist is mounted on top of the scan shield, which in turn rests on the fillet ring and the instrumented shield ring containing gamma-ray and neutron detectors. The rotational stage is shown on top of the waste can.

Using the SMS to determine the INF contents of an item is a three-part process. First, reactor physics codes use INF design and radiation information to calculate the quantities of fission products and fissile materials that should be present in the INF. Second, a radiation transport model calculates the gamma-ray and neutron response associated with the presence and quantities of these materials. Third, the SMS, properly calibrated using well-characterized INF, measures the actual gamma-ray and neutron count rates for the INF item. The agreement between the calculated and measured SMS responses provides a quantitative confirmation of the calculated nuclear material quantities. In using both physics analyses and direct radiological measurements, the SMS uses all available information to confirm fissile material inventory.

The SMS, designed to measure INF located in dry storage locations, consists of a base shield ring, an instrumented shield ring, a transitional shielding piece or “fillet ring,” and a scan shield with a top-mounted hoist (**Figure 1**). The SMS mates with a storage location and measures the INF item as it is being withdrawn from storage, through the instrumented shield ring, and into the scan shield. The SMS will be used primarily at

Argonne West's Radioactive Scrap and Waste Facility (RSWF) where spent fuel is stored in thin-walled waste cans placed in underground steel tubes 16 inches in diameter and 12 feet in length. The SMS is sized to handle top-shielded RSWF waste cans up to 12.75 inches in diameter and 75 inches long. Once a waste can is drawn into the SMS, an operator uses a hoist to position the can axially using an axial-position transducer and computer position display. The waste can is then rotated inside the SMS during the measurement by a rotational stage positioned on top of the waste can and connected to the waste can by a vacuum torque coupler. The rotation averages the gamma-ray and neutron emissions during measurement.

The SMS development, functional tests, and basic calibration measurements are anticipated to be completed by the end of FY 2000. Future work will include reactor physics modeling of INF types, the addition of more detectors to measure neutron coincidence, the addition of high-resolution gamma-ray measurements, and the inclusion of active interrogation. These additional capabilities will allow INF parameters such as fissile contents, spontaneous fission isotope contents, and burnup to be determined independently of reactor physics calculations for certain INF types and thus further confirm the validity of the calculations.

Emergency Exit Radiation Monitor

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Emergency exits are essential for all occupied buildings. The Emergency Exit Radiation Monitor (EERM) help ensures that no one can remove nuclear material under the cover of an emergency evacuation at a DOE nuclear facility.

The system is designed around a set of very sensitive sodium iodide detectors having moderate energy resolution. If any gamma emission above natural background is detected at an emergency exit (while the door is open), an alarm is generated and an energy spectrum is simultaneously collected from all detectors. The output of a video camera is also collected and stored for a time period from approximately 30 seconds prior to the onset of the alarm to 30 seconds after the alarm ceases.

The energy spectrum, stored on a separate PC, can be viewed by security personnel to determine the nature of the nuclear material exiting the facility. If the alarm was triggered by a person who has undergone radioactive-isotope treatment, this fact may be discovered from this spectrum as long as enough data was collected during the event.

The detectors are connected to each other and to the central monitor by a LONWORKS communications protocol using a single twisted-pair wire. This means of communication greatly minimizes installation costs and can interconnect detectors located up to 1,000 feet apart. The system constantly polls each detector to determine its operating fitness and notifies the central monitor if a failure occurs.

Central monitoring is fulfilled by a standard PC fitted with a LONWORKS communications interface card and a video frame grabber (**Figure 1**). This technology reduces capital and maintenance costs and permits easy upgrades. The frame grabber and camera are commercial

Figure 1. EERM display showing alarm, video image, floor plan, and spectra information.

devices and both are low cost. The central monitor can communicate directly with the existing security system at a facility.

The detector assembly is a compact, highly integrated design. It consists of an integrated high-voltage power supply, a multi-channel analyzer to collect an energy spectrum, a microprocessor, and a LONWORKS communications adapter mounted directly to the photomultiplier tube of the detector. This approach reduces manufacturing costs, reduces size, and improves reliability by eliminating almost all internal cabling. The detector assembly is mounted in a rugged, sealed (waterproof) aluminum case with lead shielding (collimation), and thermal insulation (**Figure 2**). The case's design reduces background radiation levels, consequently increasing the sensitivity of the device. It also protects it from the environment. When installed, it is further protected from collisions from machinery such as forklifts.

The first installation of this system is expected to take place at the Device Assembly Facility located at the Nevada Test Site. At this site, the detectors will be mounted inside the facility near emergency exit doors (**Figure 3**). The alarm generated from this system will be transmitted to the existing security system as well as to the EERM central monitor. The facility security system will also be notified in the event of a detector failure.

Figure 2. EERM detector.

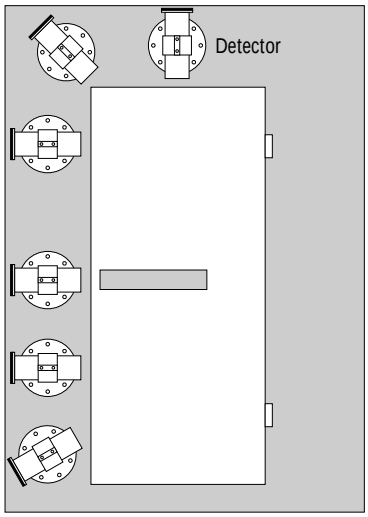

Figure 3. Sketch of an EERM installation.

ASTM Standards for Vehicle Portal Monitors

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The security plans of all DOE facilities require monitoring pedestrians and vehicles for special nuclear material (SNM) as they depart protected areas. Controlling the movement of nuclear materials is the most effective nonproliferation measure. Automatic pedestrian and vehicle portal monitors have been a very significant component of safeguards and security at U.S. nuclear facilities for 15 years. This technology, developed at Los Alamos National Laboratory, provides the outermost layer of protection against the theft of SNM.

Automatic monitors sense the presence of SNM by comparing the gamma-ray and neutron intensity of the portal while it is occupied, either by a person (**Figure 1**) or a vehicle (**Figure 2**), to an alarm threshold, which is continuously updated while the unit is unoccupied. Although these units are commercially available, they have been tailored to the requirements of each facility. Historically, Los Alamos has helped individual facilities to tailor their portals for their needs, including developing written procedures for hand-held-monitor techniques as (**Figure 3**).

The sensitivity of automatic monitors depends on many parameters, including ambient background radiation intensity, the size of the occupancy space, detector response to emitted and scattered radiation, source shielding, available monitoring time, and an acceptable nuisance alarm rate. Although a vast number of radiation monitors are commercially available, only a very few have the required SNM sensitivity and false-alarm rate. Most lack the ability to detect the low-energy gamma rays from HEU or have a greater than the desired one-per-one thousand nuisance alarm rate.

Figure 1. An automatic pedestrian SNM monitor.

Figure 2. An automatic vehicle SNM monitor.

Los Alamos has published a series of application guides, and together with the American Society for the Testing and Materials (ASTM), has written standards for testing and evaluating hand-held and pedestrian monitors. These guidelines ensure that automatic monitors are sufficiently sensitive to SNM to constitute a significant deterrent to theft. A standard for vehicle monitors needs to be written to match the stringent requirements described for pedestrian monitors. Vehicle monitors are especially difficult to evaluate because of the variance of gamma-ray shielding inherent with each vehicle. A standard that describes the procedures for evaluating vehicle monitors using SNM under realistic operating conditions would increase the effectiveness of these monitors.

A vehicle standard is especially important because the use of vehicle monitors is expanding beyond nuclear facilities to international borders. Pedestrian and vehicle monitors are currently deployed at an international border crossing. This test installation presents real-world challenges such as the innocent alarms caused by the natural radioactivity associated with shipping large quantities of produce and fertilizers.

Figure 3. Searching for hidden SNM using a hand-held radiation monitor.

Standard Options for Safeguards Seals: Replacing the E-Cup

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Going into FY 2000, Sandia National Laboratories was told by DOE that American Casting & Manufacturing Co. had sold its stamp and dies for manufacturing the E-cup loop seal [Type E seal] to the Former Soviet Union. The company was no longer manufacturing the E-cup. Because of the sale, DOE expressed its concern to Sandia about the continued use of the E-cup, which has been the mainstay loop seal in the DOE complex for the last thirty years. For FY 2000, Sandia was tasked to find loop seals that could replace the E-cup.

The seals are being evaluated for DOE's "two-person rule" environment. This rule states "at any time there is access into a security vault, there must be two or more people within eyesight of each other at all times within the vault." The goal of these evaluations is to find loop seals as tamper-resistant as the E-cup.

Sandia has identified five loop seals that are now being evaluated ("blackhatted") by its surety development group: (1) Toolless Roto-Seal (E.J. Brooks, **Figure 1**), (2) Quick Seal (NIC, **Figure 2**), (3) PSW97 (American Casting and Manufacturing, **Figure 3**), (4) Clip-Lok (E.J. Brooks, **Figure 4**), and (5) Clear Guard (RELCOR, **Figure 5**). These loop seals are listed in the order of priority given them by Sandia's surety development

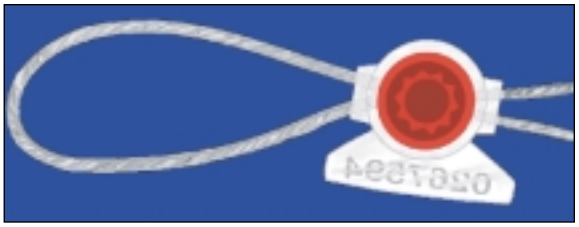

Figure 1. Toolless Roto-Seal (E.J. Brooks).

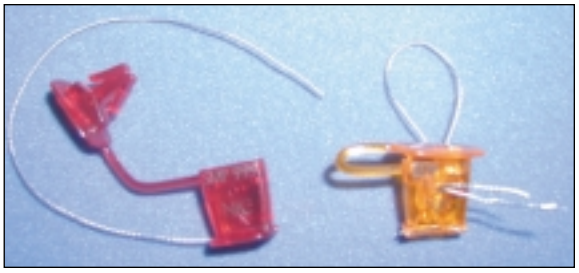

Figure 2. Quick Seal (NIC).

group. Los Alamos National Laboratory and Pantex have expressed interest in having Sandia evaluate NIC's Quick Seal because Pantex currently uses it. Los Alamos is considering this seal as the replacement for the E-cup. Los Alamos is also very interested in E.J. Brooks' Toolless Roto-Seal, and because they are quickly running out of their E-cup inventory, they have requested we accelerate our evaluation.

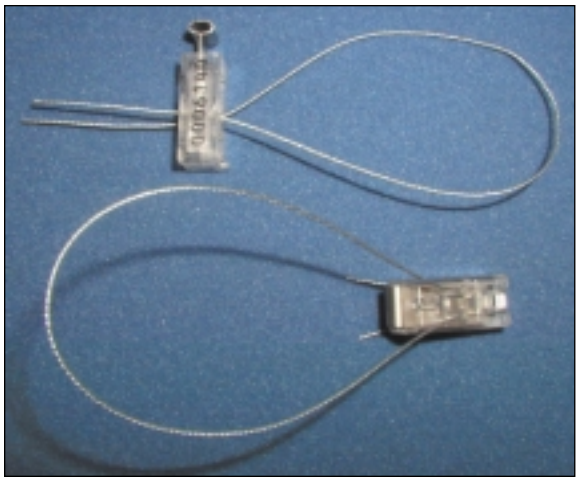

Figure 3. PSW97 (American Casting and Manufacturing)

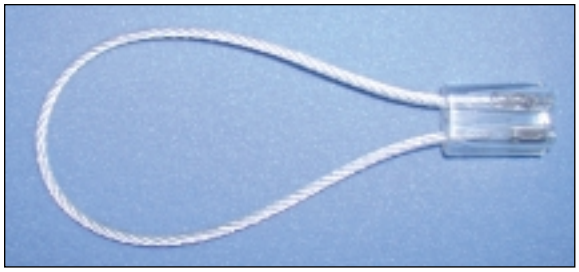

Figure 4. Clip-Lok (E.J. Brooks).

Figure 5. Clear Guard (RELCOR).

Fiber-Optic Seals: Tamper Resistance and Active Seals

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Tamper-Resistance

In late July of FY 1999, DOE requested recommendations for passive loop seals with higher tamper-resistance times than those in use throughout the DOE complex. After reviewing the more widely used loop seals, Sandia National Laboratories recommended Aquila Technology's Cobra fiber-optic seal as having the highest tamper-resistance time. This seal costs $30 for the seal and $2 per meter for the fiber-optic loop; the digital imaging device cost $15,000. Sandia felt the next best loop seal was a more realistic choice within the DOE complex: the E.J. Brooks' Fiber-Lok, around $6.50 per seal (12-inch fiber-optic loop) and a Polaroid camera costing $1,500 (**Figure 1**).

Figure 1. E.J. Brooks' Fiber-Lok and Polaroid camera. The Polaroid camera takes a picture of the seal.

Sandia agreed to procure the Fiber-Lok but found only one facility interested in evaluating the seal. These data will be used to generate a report. That facility is the Canister Storage Building currently under construction within the Hanford reservation. The Fiber-Lok could secure the exposed tops (**Figure 2**) of each Multi-Canister overpack storage tube. This will tamper-protect the Category II quantities of spent nuclear fuels. If the concept is accepted and schedules go as planned, the Fiber-Loks will be installed after the facility is opened (end of 2000–first quarter 2001).

Active Seals Pilot Projects at Hanford and Los Alamos

Sandia was asked in FY 1999 to identify DOE facilities which might be interested in active seal technology to increase tamper-resistance time in one of their vaults. The project would determine the cost–benefit tradeoffs of implementing such technology, including the installation costs. Two sites expressed an interest: 200 West at the Hanford site and Area TA-55 at Los Alamos National Laboratory.

In the 200 West facility's vault complex, the Fiber SenSys system was installed in a cubicle-array room (class V doors) located within a vault complex also with class V doors (**Figure 3**). At Los Alamos, in a temporary metal building, Fiber SenSys will be evaluated to determine if it can adequately protect waste storage drums (**Figure 4**). Waste waiting to be transported to the Waste Isolation Product Plant facility will be stored here.

Figure 2. The Fiber-Lok loop seal is criss-crossed over tops of the Multi-Canister overpack storage tube.

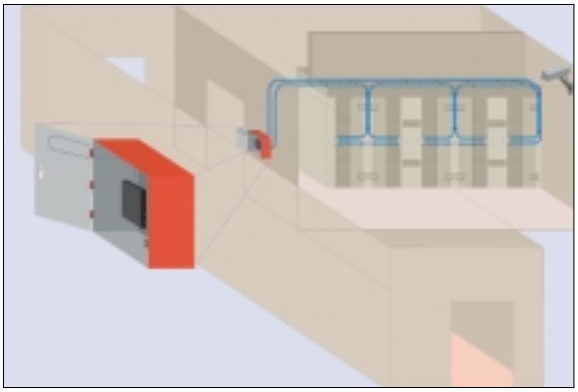

Figure 3. Located at the 200 West facility on the Hanford site, the Fiber SenSys is being evaluated in a vault room within a vault.

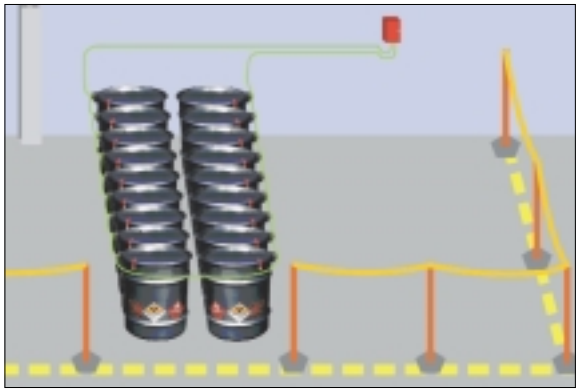

Figure 4. The Fiber SenSys can protect the stored waste and the empty 55-gallon drums (DOE requirement).

SNM Container Monitoring System

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The long-term inventory of special nuclear materials (SNM) to provide materials protection control and accountability is of the highest importance both nationally and internationally to verify treaty compliance. Physical and statistical methods currently used to inventory SNM are costly and account for only a small percentage of the total number of containers in storage.

A self-contained, self-powered sensor suite to measure inside a sealed SNM container will provide data for evaluating and verifying the physical condition and location of the container. The technology can be implemented without breaching or modifying the SNM container. Measurement data along with a unique container identification code can be automatically accessed via a wireless radiofrequency (RF) tag to assess the inventory remotely and report the status of long-term storage and even during container transport (**Figure 1**). Automated data acquisition will greatly reduce the need for manual audits and radiological evaluations to determine container integrity. This will result in reduced personnel radiation exposure, improved data quality, and an accurate historical record on each container.

We demonstrated the system in the laboratory using an AT400A SNM container. The container's internal electronics suite consisted of thermo-electric generators (TEG), a temperature sensor, a micro-controller, and an acoustic data link. On the container's external wall was an acoustic datalink and RF tag to transmit the internal sensor data to a remote inventory-control station (**Figure 2**). Using the heat produced by the SNM, the thermo-electric generator powers the sensor, micro-controller, and

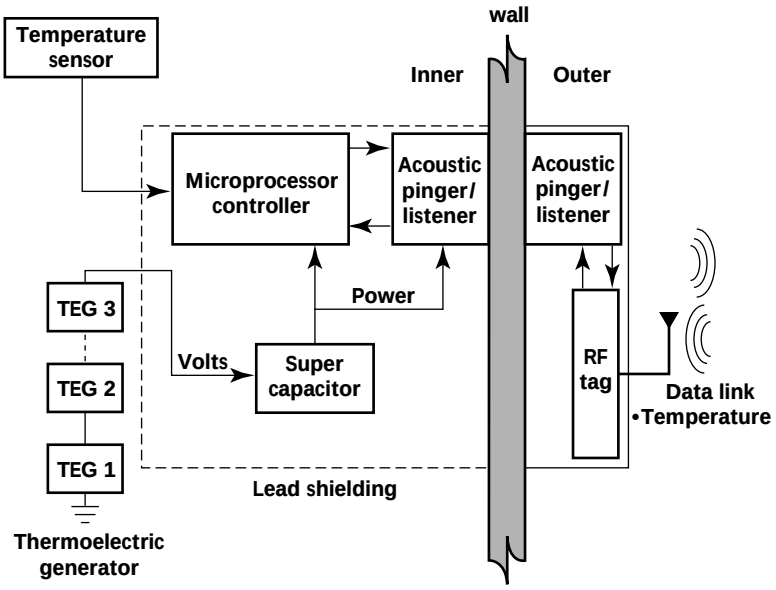

Figure 1. SNM container monitoring system block diagram.

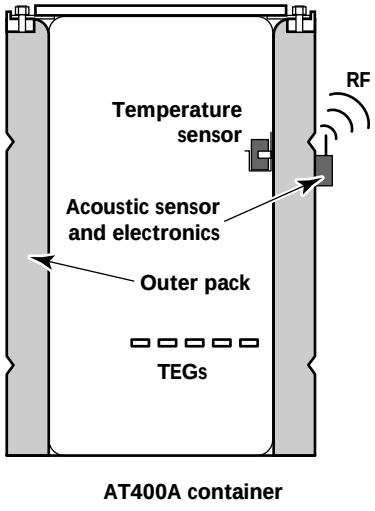

Figure 2. AT400A container.

acoustic data link indefinitely. Internal sensor data are transmitted to the outside without breaching the container via acoustic data link. The sensor data are tagged with a unique SNM container ID number and transmitted via an RF link to the inventory-control station where the data are processed, analyzed, and reported automatically (**Figure 3**).

During a typical operation, a background timer periodically prompts for a sensor-suite measurement and a data transfer. The data transfer is done acoustically by employing a pinger–listener technique. High-frequency sound (~400 kHz) is propagated through the inner container wall and coupled via an acoustic waveguide to the outer shipping container and then through its wall to the outside wireless RF tag. The acoustic signals are modulated, transmitting data in a digital format. The minimum power required to operate the internal sensor suite electronics is 1 volt. The TEGs will be strategically located to optimize the heat generated by the SNM to achieve the electrical power necessary to operate the sensor suite. SNM container monitoring also can be measured via remotely located host requests, as the acoustic modem is bi-directional and can send commands to the internal electronics.

Both the internal sensor suite and outer wireless RF tag packages are miniature (~2 inches × 1 inch × 0.3 inch). The outer wireless RF tag can be powered from a battery or a low-voltage DC power source. Performance tests on an AT400A container have shown that a simulated internal heat source could indefinitely support measurements on a 5-second update basis.

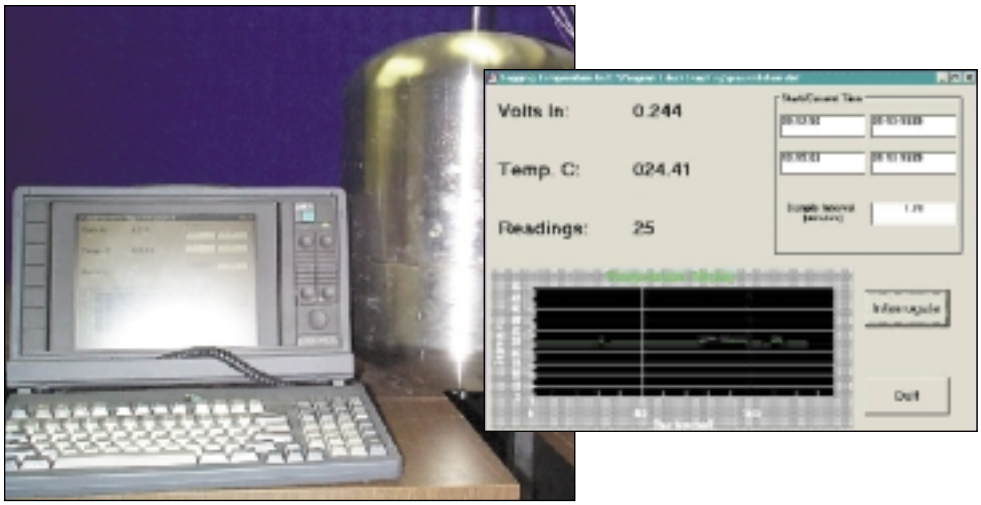

Figure 3. Remote inventory control station and measurement report.

Multi-Instrument Collect (MIC)

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Figure 1. Multi-Instrument Collect main menu depicting support for different instrument types.

Currently, various standardized instruments accomplish specific NDA tasks. Each of these instrument types is unique as to its configuration, data acquisition, status monitoring, and data retrieval. This diversity creates problems in setup, maintenance, use, and troubleshooting the instrument as well as training end-users. The Multi-Instrument Collect (MIC) program, created by Los Alamos National Laboratory, addresses most of these issues.

The MIC program collects and archives data simultaneously and without an operator present from multiple, distributed data-acquisition instruments, supplying a consistent interface for all the instruments. It can collect from up to 50 instruments and communicate on up to 30 channels. **Figure 1** shows the main menu of MIC configured to support five instruments of four different types: ISR, GRAND3, Mini-Grand, and an uninterruptable power system. MIC also exports collected data to removable disks.

The MIC system consists of one main program, MIC, and a collection of support programs designed to accomplish various tasks associated with collecting data but not associated with a specific instrument or instrument family. Designed to run on a multi-processor Windows NT®-based system, it can run adequately on a single-processor platform as slow as 133 MHz. The interface philosophy is called "Status-at-a-Glance" or StaG (**Figure 2**). Bold characters convey information that changes; pictographs or icons for status information at a high level, colors on button faces and text reinforce rapid communication to the user, and data are presented in a hierarchical structure to minimize overload and confusion.

MIC supports the GRAND3, Mini-Grand, Intelligent Shift Register, Advanced Multiplicity Shift Register, Multi-Channel Analyzer/1KADC, and the Jomar JSR-12. Because MIC is written in C++ using industry-standard object-oriented programs, the expense of adding support for other existing or new instruments is minimal.

MIC runs unattended on a PC with one or more Pentium processors running the Windows NT® 4.0 or above operating system. The program collects data via one or more Intelligent Local Operating Network (ILON), ILON serial extensions, or serial communications lines with Ethernet being added. Data collection is automatic and continues until an operator intervenes. All raw data are stored in files on local or remote disk drives or RAID systems. Critical events and maintenance files are also created. Each record is time-stamped.

MIC transmits state-of-health data in real time and in near real time to multiple receivers via an attached network. The network may be connected to an internet or intranet, allowing monitoring anywhere in the world or it may be deactivated completely. Multiple techniques are available in MIC to meet nearly any system requirement.

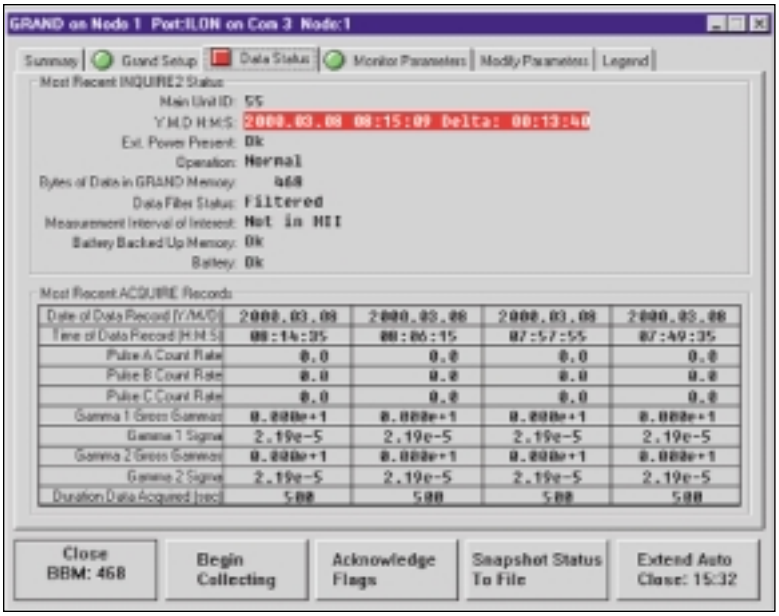

Figure 2. An example of a Data Status page from the MIC instrument support object for a GRAND3. These screens are very similar to the associated pages on each of the other instrument types supported.

Integrating NDA Instruments with MC&A Systems

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The nondestructive assay (NDA) software developed over the years was designed using different commercial libraries, tools, and databases. The hardware and software tools needed to develop the NDA software are no longer supported or no longer available. Older NDA systems were designed around computers with memory and data storage limitations. Data were analyzed and the results stored in data files or databases. Because of storage limitations, much of the raw data could not be stored. Advances in computers and operating systems allow us to design new NDA instruments with the ability to store large amounts of data to databases locally or over the network using standard software tools. Unfortunately, older NDA systems have not benefited from these advances.

A complete software redesign for all NDA software is not possible. Instead, we have designed a cost-effective solution to allow NDA instruments running older software to save any data to a file and have that file automatically sent to a local or remote Microsoft Access® database. This is done using an NT Service designed for this purpose. The service checks for files in the appropriate directories and automatically transfers them to a database. The service mechanism provides an elegant way to create background tasks for NT machines. Once the service is installed, it is started automatically when the machine is booted up. The service can also be controlled from the Windows NT® "Control Panel."

The database and its location are specified using Microsoft's ODBC data source administrator. The ODBC interface enables applications to access data in database management systems that use Structured Query Language (SQL) as a data access standard. The software has been tested to access databases located on the NDA application computer and over the network.

Many of the NDA instruments collect spectral data from multi-channel analyzers (MCA). Storage of raw spectral data to an Access database is difficult because the Access database was not designed to store large arrays of data. By using some of the same structures that Access uses to store documents, we can store raw spectral data. **Figure 1** demonstrates this code being used by a solution assay instrument to retrieve previous assay spectrum data.

By modifying the existing NDA software to store raw data into a common database, we provide a better trail for diagnosing instrument measurement failures. By saving the raw measurement data, it may be possible to track trends in the measurement data and to later reproduce and verify results. Once the data are in the Access database, we can easily examine the data and generate reports using simple, built-in industry-standard tools. **Figure 2** shows how standard Microsoft Access® tools represents data from a segmented gamma scanner.

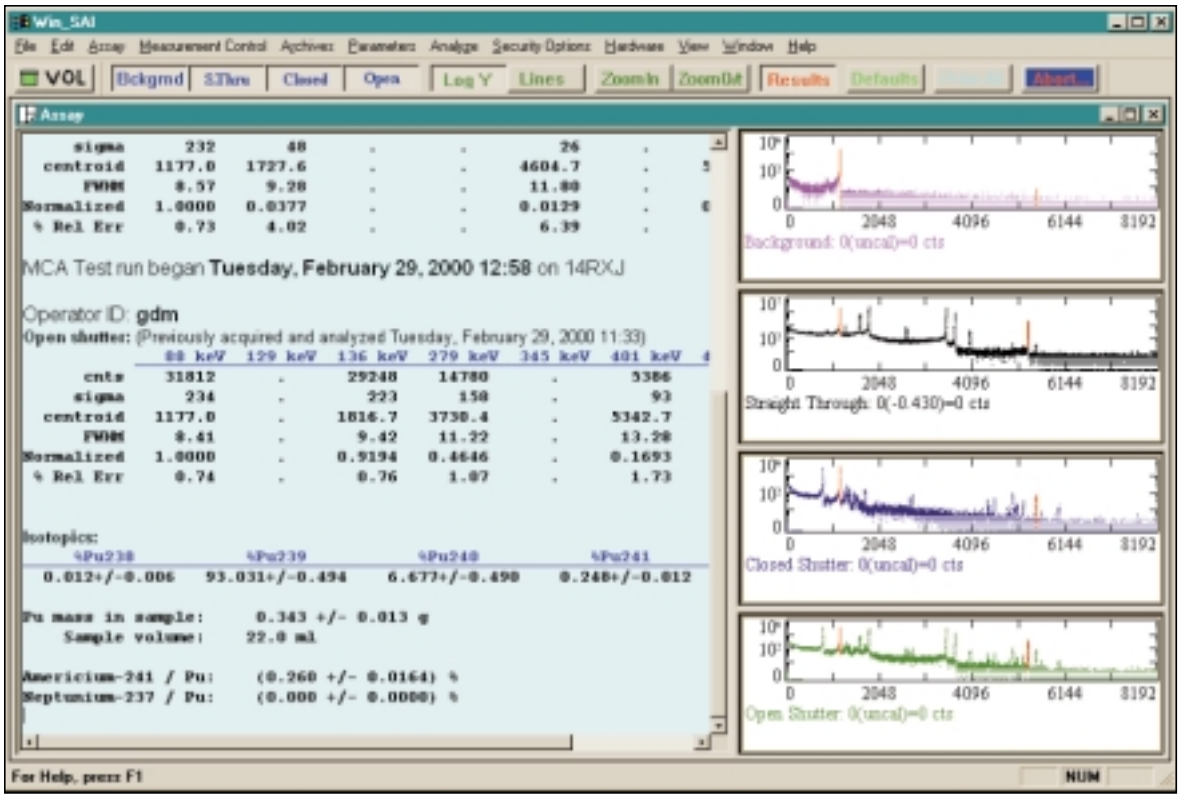

Figure 1. Reading previously stored spectrum data.

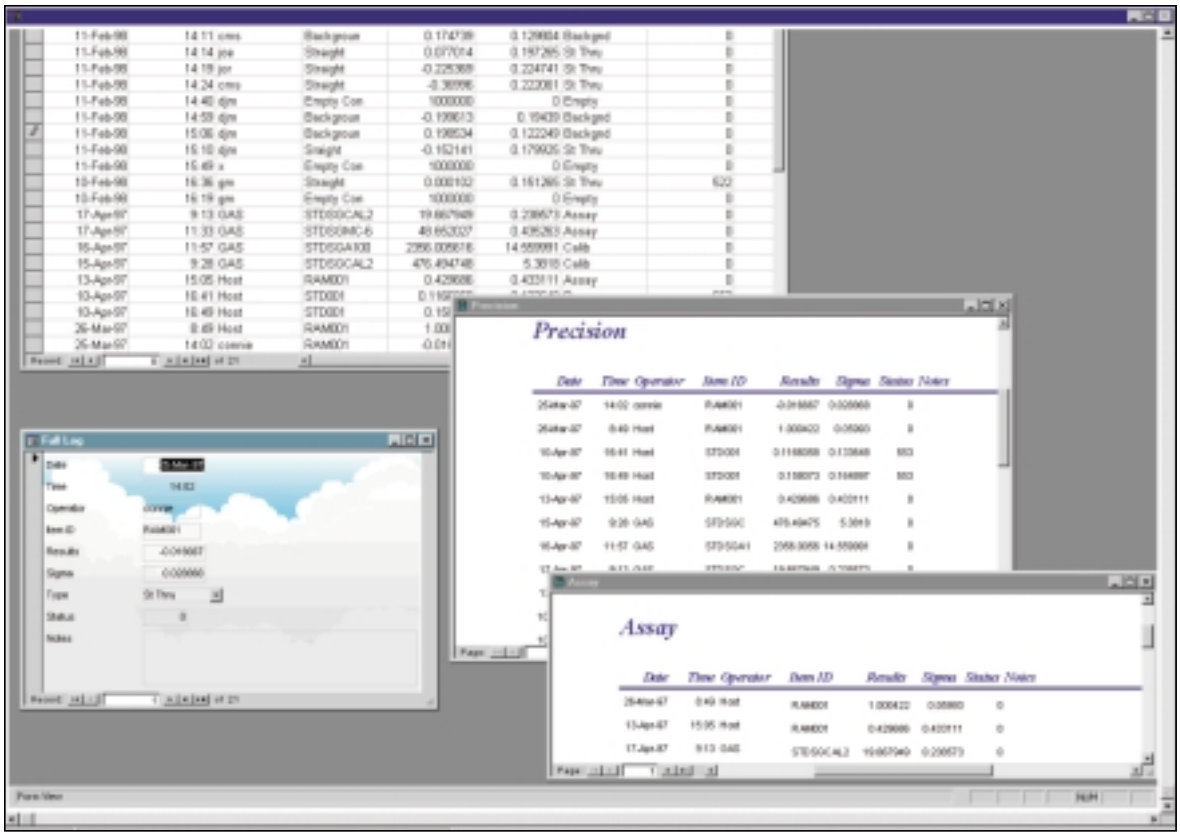

Figure 2. Using standard Microsoft tools to display saved data.

Interface and Data-Transfer Standards Between Safeguards Instruments and Computerized MC&A Systems

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Standardization and modularization of the interface between safeguards instruments and MC&A databases are required to implement a uniform data construct throughout the DOE complex. In this project, a standard for data transfer between nuclear-material-measurement instruments and databases that track the materials will be developed. The standardization effort will focus on data collection, data validation, data transfer, automated entry into MC&A database, and database design and implementation that facilitate automated transfer. These standards will be developed via the formation of a working group. The stakeholders in this process include DOE MC&A management, site MC&A organizations, site MC&A information systems operations, DOE MC&A information systems developers, DOE and commercial instruments developers.

Currently, measurements or analysis performed at facilities require that analysts review and validate the measurement or analysis, provide reports (manually) to the nuclear materials management organization, and enter data into the MC&A database. Making better use of available technology, the analysis would be performed, analysts would review and validate the measurement or analysis on-line, transactions would be sent to the MC&A database by keyless automated entry into the database. This capability could provide inventory data faster with less labor involved in getting data to database and no manual data entry (less errors, better quality assurance). These standards will also ease the integration of instruments and databases from different sources (facilities/laboratories/commercial providers). It will also ease the modification of instrumentation or information systems.

Data collection needs to be performed in a way that meets the data-transfer requirements so there is a minimum amount of information requiring collection and storage or translation into a format consistent with

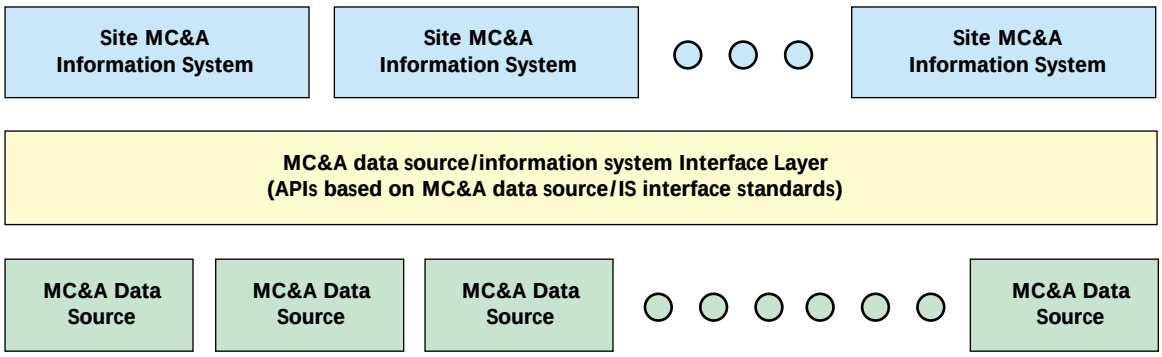

Figure 1. Architecture for the transfer of data to MC&A systems.

the data-transfer requirements. This effort might also touch on file-format standards and standards for instrument application programming interfaces (APIs). An initial direction for the standard might be for a readily divisible, text-based format.

Two options for implementing a validation methodology are currently being considered. First, analysts validate data at the instrument. Second, the data is transferred to the MC&A Information System (IS) as a transaction and validated within the IS before automated entry into the database.

Two issues for data transfer are protocol (data format) and content. Because different facilities have different data needs, the working group will define a kernel or minimum set of transferred data. The data-transfer format must be extensible to handle this kernel but still permit various facility-specific data fields.

Automated entry will be facilitated by a standard API (**Figure 1**) for the MC&A information system. Like the data-transfer format, this standard must be extensible to handle facility-specific needs. This API, although flexible in nature, will be specifically targeted for entry of data into LANMAS systems, the DOE-adopted standard computerized accounting system.

Aspects of database design and implementation can help or hinder automated transfer and the integration of subsystems of different sources. Standards or guidelines will be discussed on this topic. Because the DOE standard for MC&A information systems is LANMAS, discussion will focus on implementation around LANMAS.

The working group will consider standards and guidance for existing instrumentation and upgrades as well as new instruments. At the other end, standards and guidance will be generated for existing and new MC&A database systems.

DOE laboratories implementing technologies to manage unique inventory and radiation detection needs at each facility

Lawrence Livermore National Laboratory

- Segmented gamma barrel scanner
- Shuffler
- Calibrating Pu standards
- Calorimetry
- Training
- Neutron multiplicity counter
- Isotopic analysis–MGA
- Tomography
- Signal processing
- Portable spectroscopy systems
- In-vault measurements
- Isotopic analysis of weapons' aging

Oak Ridge National Laboratory

- Wireless sensors
- Segmented gamma barrel scanner
- Training
- Calibrating U standards
- Nuclear Materials Identification system (NMIS)

Nevada Test Site

- Isotopic analysis–MGA
- Add-a-source
- High-energy neutron counter
- Segmented gamma barrel scanner
- Training

New Brunswick Laboratory (Argonne)

- Segmented gamma barrel scanner
- NDA standards report

Los Alamos National Laboratory

- ARIES NDA
- Calorimetry
- Neutron coincidence counter
- Isotopic analysis–PC/FRAM
- High-energy neutron counter
- Add-a-source
- Vault monitor
- Calibrating Pu standards
- Segmented gamma barrel scanner
- Hybrid densitometer
- Solution assay instrument
- Anomaly detection
- Portal monitors
- Hand-held monitors
- CTEN

Sandia National Laboratories

- Segmented gamma barrel scanner
- LANMAS
- Training
- Material Monitoring System
- Track Right
- Strategic Reserve Stage Right
- Radiation Physical Inventory Pallet (RPIP)
- Seals testing and selection

Waste Isolation Pilot Plant (WIPP)-related

- Training
- Tomographic gamma scanner
- Segmented gamma barrel scanner
- CTEN
- High-energy neutron counter
- Monitoring

Savannah River Site

- Neutron multiplicity counter
- Pu heat-source isotopic assay system
- Image processing
- Calorimetry
- NDA standards report
- Training
- LANMAS
- Neutron coincidence counter
- MAWST
- Segmented gamma barrel scanner
- Calibrating Pu standards
- Barrel scanner analysis and control
- Isotopic analysis–MGA and PC/FRAM

Idaho National Engineering and Environmental Laboratory

- PINS
- NDA standards report
- Image processing
- Isotopic analysis–MGA and PC/FRAM
- Segmented gamma barrel scanner
- Neutron coincidence counter

Hanford

- NDA standards report
- Isotopic analysis–MGA
- Segmented gamma barrel scanner
- Calibrating Pu standards
- Neutron coincidence counter
- Training

Rocky Flats Environmental Technology Site

- Neutron multiplicity counter
- Training
- Holdup technologies
- Tomographic gamma scanner
- Segmented gamma barrel scanner
- Calorimetry
- Isotopic analysis–PC/FRAM
- Calibrating Pu standards
- Image processing
- Portal monitors
- Hand-held monitors

Special Technologies Laboratory (Bechtel)

- Pocket pager

Y-12 National Security Complex

- Holdup technologies
- CWAM
- Shuffler
- Training
- CAVIS monitoring
- SmartShelf
- ReflectoActive seals
- Nuclear Materials Identification System (NMIS)
- Segmented gamma barrel scanner
- Segmented gamma can scanner
- Solution assay instrument
- Active well coincidence counter
- Portal monitors
- Hand-held monitors

Meeting interagency needs for the U.S. Government and policy requirements

U.S. Customs

- Field radionuclide identification system
- Pocket pager
- Portal and hand-held monitors
- Active Interrogation Package Monitor (AIPM)

Russian Projects, IAEA Bilateral Cooperation

- Isotopic analysis–MGA and PC/FRAM
- Electromechanically cooled gamma-ray detectors
- Portable uranium isotopic analysis systems
- High-energy neutron counter
- Hybrid densitometer
- INCC software
- Portals
- Image processing
- NDA standards report
- Training
- Neutron coincidence counter
- ARIES
- Segmented gamma barrel scanner
- Calorimeters
- Computerized MC&A
- Active Interrogation Package Monitor (AIPM)
- Hand-held monitors
- Portable XRF–solution assay

Licensing applicable technologies to speed distribution throughout the DOE complex

Corporate Partners

- Isotopic analysis–MGA and PC/FRAM
- Electromechanically cooled gamma-ray detectors
- Digital signal processing to increase sensitivity
- Portable uranium isotopic analysis systems
- High-energy neutron counter
- Calorimeters
- Hand-held monitors
- Portal monitors
- Multi-channel analyzers
- Tomographic gamma scanner
- Holdup technologies

FY 2001 MC&A Projects: New Starts

MGA Analysis of Elevated Concentrations of ^{238}Pu in Plutonium. This project will provide a means to assay materials that contain elevated (>1%) ^{238}Pu that result from the accelerated aging of plutonium materials. Current assay capabilities cannot meet this requirement.

Development of Calibration Techniques for Pan Shuffler. This project will calibrate uranium oxide in the Passive–Active Neutron (PAN) shuffler to measure uranium oxide items of variable enrichments as well as uranium oxide in mixed-oxide fuels for all inventory items requirements per Lawrence Livermore's MC&A plan.

Advanced Inventory Methodology. This project will establish technical criteria to evaluate candidate approaches to continuous inventory in terms of (1) the ability to meet inventory objectives and (2) the effects on other safeguards elements including inventory-difference evaluations.

Compact Spectroscopic Imaging for Facility Safeguards in Non-Static Material Balance Areas. This project will develop and test analysis algorithms and methodology applied to gamma-ray data from commercial equipment to assay the nuclear material *in situ* and determine its distribution.

Accurate MEGA/RAM for Accountability of Holdup and Criticality Safety. (MEGA/RAM = Measure and Evaluate Generalized geometry–gamma Assay/Rapid Automated Method). This project will produce a rapid, automated system for determining accurate holdup quantities plant-wide and semi-quantitatively screen deposits for criticality safety.

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About the Technology Development Program

The Technology Development Program (TDP) is a critical element within DOE's Office of Safeguards and Security. The TDP is the only technical program within the DOE focused on developing technologies to protect nuclear weapons, special nuclear materials, and other high-value assets. Programmatically, the TDP collects "user needs" from DOE's field elements, canvasses industry and other federal agencies for solutions, and formulates technology development projects to address requirements for which no available solution currently exists. TDP technologies are diverse to counter the threats posed by sophisticated insiders, terrorist teams, explosives, computer hackers, and emerging threats such as chemical agents and directed-energy weapons. Together with the national laboratories, the TDP is making a difference in our national security.

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